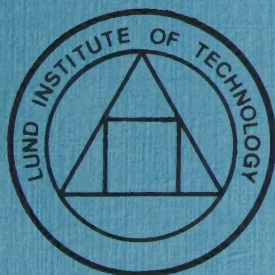


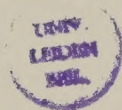
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TIME RESOLVED LASER
SPECTROSCOPY OF RYDBERG STATES
IN GROUP IIA AND IIIA ELEMENTS

GÖRAN JÖNSSON



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Using laser techniques in connection with atomic beams selective excitation of Rydberg states was performed. Most of the studied states were populated with two-step excitation. Some states were reached directly from the ground state or from a metastable state and occasionally two-photon absorption was used. The excitation sometimes involved the use of frequency-doubling and Raman-shifting techniques.

By time-resolved detection of the decay light from an excited state, primarily information on the radiative lifetime is obtained. With two different experimental arrangements radiative lifetimes in several Rydberg states have been determined. Using the quantum-beat technique time-resolved detection also provides information on energy splittings. This technique was used to measure hyperfine-structure splittings. To obtain information about hyperfine-structure constants and isotope shifts high-resolution spectroscopy on a collimated atomic beam was also employed.

The behaviour of highly excited alkali atoms is mostly hydrogen-like and well predicted by theory. Group III A elements have one electron outside a closed subshell and are expected to behave like alkali atoms. The situation is however complicated by the presence of doubly excited states. Configuration interaction can then cause a partial mixing between singly and doubly excited states. In the alkaline-earth atoms (group II A elements), having two electrons outside closed shells, the situation is similar. In a Rydberg series of one excited electron the admixture of doubly excited states can cause large deviations from hydrogenic behaviour. Another complication in the alkaline-earth atoms is the mixture of singlet- and triplet states.

The presented results from measurements made on Na, Mg, Ca, Sr, Ba, Al, Ga and In will primarily serve as a basis for tests of modern atomic-physics models. Some of the lifetime values are also of astrophysical interest.

Key words Laser spectroscopy, Rydberg states, time resolved, high resolution, two-step excitation, metastable state, two-photon absorption, frequency doubling, Raman shifting, radiative lifetime, quantum beat, hyperfine structure, isotope shift, group IIA and IIIA

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TO MATINČEN AND MY FRIENDS



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Keywords: Laser spectroscopy, Rydberg states, time resolved, high resolution, two-step excitation, metastable state, two-photon absorption, frequency doubling, Raman shifting, radiative lifetime, quantum beat, hyperfine structure, isotope shift, Stark shift, alkali atoms, group III A elements, alkaline-earth atoms, configuration interaction, singlet-triplet mixing.

This thesis is based upon the following papers.

1. Natural Radiative Lifetimes in the Interacting $5snd\ 1,3D_2$ Sequences in Sr.
P. Grafström, Jiang Zhan-Kui, G. Jönsson, C. Levinson, H. Lundberg and S. Svanberg.
Phys. Rev. A 27, 947 (1983).
2. Natural Radiative Lifetimes in the $1P_1$ and $1F_3$ Sequences of Sr I.
G. Jönsson, C. Levinson, A. Persson and C.-G. Wahlström.
Z. Phys. A, in press.
3. Natural Radiative Lifetimes and Stark-Shift Parameters in the $4p^2$ Configuration in Ca I.
G. Jönsson, C. Levinson and S. Svanberg.
Physica Scripta, in press.
4. Natural Radiative Lifetimes in the $3sns\ 1S_0$ and $3snd\ 1D_2$ Sequences of Magnesium.
G. Jönsson, S. Kröll, A. Persson and S. Svanberg.
Submitted to Phys. Rev.
5. Natural Radiative Lifetimes and Hyperfine Structure of the $4s^2 6p\ 2P_{3/2,1/2}$ Levels of Gallium.
G. Jönsson, C. Levinson, S. Svanberg and C.-G. Wahlström.
Phys. Lett. A 93, 121 (1983).
6. Lifetime Measurements in the $2S_{1/2}$ and $2D_{3/2,5/2}$ Sequences of Indium.
G. Jönsson, H. Lundberg and S. Svanberg.
Phys. Rev. A 27, 2930 (1983).
7. Natural Radiative Lifetimes in the $2S_{1/2}$ and $2D_{5/2,3/2}$ Sequences of Aluminum.
G. Jönsson and H. Lundberg
Z. Phys. A. - Atoms and Nuclei 313, 151 (1983).

8. Hyperfine-Structure Study of the 7, 8 and 9 $2p_{3/2}$ States in ^{23}Na Using Quantum-Beat Spectroscopy.
Jiang Zhan-Kui, G. Jönsson and H. Lundberg.
Physica Scripta 26, 459 (1982).
9. Hyperfine Structure and Radiative Lifetimes in the $3s^2np$ $2p_{3/2}$ Sequence of ^{27}Al using Time Resolved Laser Spectroscopy.
G. Jönsson, S. Kröll, H. Lundberg and S. Svanberg.
Z. Phys. A, in press.
10. Hyperfine Structure and Isotope shift of Highly Excited Barium-I States.
P. Grafström, Jiang Zhan-Kui, G. Jönsson, S. Kröll, C. Levinson, H. Lundberg and S. Svanberg.
Z. Phys. A - Atoms and Nuclei 306, 281 (1982).

I. ATOMIC STRUCTURE

I.1 Introduction

Around the turn of this century one of the unsolved physical problems was the explanation of emission spectra coming from free gas atoms. J.J. Balmer found regularities in the hydrogen-atom spectrum and Janne Rydberg discovered a similar behaviour for highly excited alkali atoms. This inspired Niels Bohr to construct an atomic model that describes the gross structure in hydrogen. Since then a tremendous amount of spectroscopic information has been obtained. With the invention of resonance methods very subtle atomic structures could be investigated. All this information combined with results from scattering experiments has served as a basis for sophisticated atomic-physics models.

The invention of the tunable dye laser (about 15 years ago) meant an enormous progress for spectroscopy. Lasers provide very intense and narrow-band radiation that can be used in selective excitation of atomic states. New sensitive spectroscopic methods were developed and during the last decade the study of highly excited atomic states (Rydberg states) using laser techniques has been performed by several atomic physics groups. The interest was first focused on the alkali atoms. Having only one electron outside closed shells and no doubly excited states below the first ionization limit, the basic behaviour is hydrogen-like. However, even here spectacular effects like inversion of certain fine- and hyperfine structure levels can be found. These effects, particularly due to polarization of the closed electron core, are now well understood through theoretical calculations, mainly by many-body perturbation theory [1].

Group III A atoms have one electron outside a closed subshell with two s-electrons. Their behaviour is primarily expected to be alkali-like but, due to the presence of doubly excited states, large deviations can be found. Configuration interaction can cause a partial mixing between singly and doubly excited states. If states with different LS-character are mixed into a Rydberg series this is reflected in the radiative properties of the atom. The alkaline-earth atoms have two electrons outside closed

shells, and here the effects of adding an extra electron to an alkali system can be studied. The deviations from a hydrogenic behaviour in a Rydberg series can partly be explained by the admixture of doubly excited states. Mixture between singlet and triplet states is another complication.

In this work laser-spectroscopic methods have been used to obtain information on radiative properties of alkali atoms, alkaline-earth atoms (group II A elements) and group III A atoms. Using time-resolved detection after a pulsed excitation, information on radiative lifetimes has been obtained. With the same type of experimental set-up hyperfine-structure splittings were measured using the quantum-beat method. Information on hyperfine structure was also obtained using high-resolution spectroscopy on a collimated atomic beam. Apart from some radiative lifetimes that are of astrophysical interest, the presented results will primarily serve as a basis for tests of modern atomic-physics models.

The presented papers contain natural radiative lifetime measurements in Sr (1P_1 , 1D_2 , 3D_2 and 1F_3 sequence), Ca ($4p^2$ configuration), Mg (1S_0 and 1D_2 sequence), Ga ($6p\ ^2P_{3/2,1/2}$ states), In ($^2S_{1/2}$ and $^2D_{5/2,3/2}$ sequences) and Al ($^2S_{1/2}$, $^2P_{3/2}$ and $^2D_{5/2,3/2}$ sequences).

Hyperfine-structure studies have been performed using time-resolved technique in ^{23}Na ($7-9\ ^2P_{3/2}$ states) and ^{27}Al ($^2P_{3/2}$ sequence) and using high-resolution spectroscopy in Ba (1D_2 sequence) and Ga ($6p\ ^2P_{3/2,1/2}$ states).

I.2 Hydrogen-Like Atoms

In order to understand the properties of Rydberg states it is necessary to first review the spectroscopic properties of the hydrogen atom. Neglecting relativistic effects and assuming infinite nuclear mass the following properties can be deduced as a function of the principal quantum number n and the orbital angular momentum number l [2,3]:



1. Energy levels E_n are expressed by

$$E_n = - \frac{Rhc}{n^2} \quad (1)$$

where c is the speed of light, h is the Planck constant and R is the Rydberg constant.

2. The energy difference ΔE between two neighbouring states in a series is

$$\Delta E = Rhc \frac{2n+1}{n^4 - 2n^3 - n^2} \quad (2)$$

$$\Delta E \sim n^{-3}, \text{ for higher } n\text{-values} \quad (3)$$

The number of states in an energy interval is then expected to scale as n^3 .

3. The expectation value of the electron-orbit radius $\langle r \rangle$ is given by

$$\langle r \rangle = \langle nl | r | nl \rangle = \frac{1}{2} [3n^2 - 1(1+1)] \quad (4)$$

$$\langle r \rangle \sim n^2 \quad (5)$$

4. Spin-orbit interaction and hyperfine structure scale like $\langle r^{-3} \rangle$ and behave like

$$\langle r^{-3} \rangle = \langle nl | r^{-3} | nl \rangle = [n^3 l(l+\frac{1}{2})(1+1)]^{-1} \quad (6)$$

$$\langle r^{-3} \rangle \sim n^{-3} \quad (7)$$

5. The transition probability per unit time from a Rydberg state $|i\rangle$ to a low lying state $|j\rangle$ is proportional to

$$|\langle i | \underline{D} | j \rangle|^2 \sim n^{-3} \quad (8)$$

where $\underline{D}$ is the electric dipole operator.

6. The transition probability per unit time from a state $|nl\rangle$ to a neighbouring Rydberg state, for instance $|nl-1\rangle$ is proportional to

$$|\langle nl | \underline{D} | nl-1 \rangle|^2 = \frac{3}{2} n \sqrt{n^2 - l^2} \quad (9)$$

$$|\langle nl | \underline{D} | nl-1 \rangle|^2 \sim n^2 \quad (10)$$

For atoms that are hydrogen-like the same scaling laws are expected if n is replaced by the effective principal quantum number

$$n^* = n - \delta_l \quad (11)$$

The quantum defect δ_l is a function of l but constant for higher n -values in an unperturbed series. In the alkali atoms, having only one electron outside closed shells, the mentioned trends are well fulfilled. In non-alkaline systems large deviations can be expected in some energy regions. In order to fit the over-all behaviour, small modification of the powers of n^* may also be required.

I.3 Atomic Energy Structure

The energy-level structure in an atom can be expressed as eigenvalues E to the time-independent Schrödinger equation

$$H\Psi = E\Psi \quad (12)$$

where H is the energy operator (or the Hamiltonian) and Ψ is the wavefunction for the atomic system. The non-relativistic Hamiltonian can, in atomic units, be written as

$$H = -\frac{1}{2} \sum_i \nabla_i^2 - \sum_i \frac{Z}{r_i} + \sum_{i < j} \frac{1}{r_{ij}} + H_{fs} + H_{hfs} + H_{is} + H_m + H_e \quad (13)$$

where Z is the nuclear charge, r_i is the distance between the electron and the nucleus and r_{ij} is the distance between electron i and j . The summation is performed over all electrons in the atom. Each term in the Hamiltonian correspond to a physical interaction and gives a contribution to the total energy of the studied atomic system.

The first three terms represent the kinetic energy due to the motion of the electrons, the Coulomb interaction between the electrons and the nucleus and the Coulomb interaction between the electrons. These three terms give rise to the gross structure of the energy-level diagram. The fourth term represents spin-orbit interaction and causes the fine-structure splitting.

The fifth term represents the interaction between the electrons and the magnetic and electric moments of the nucleus and causes the so called hyperfine structure (hfs).

$$H_{hfs} = a \underline{J} \cdot \underline{I} + b \frac{6(\underline{J} \cdot \underline{I})^2 + 3(\underline{J} \cdot \underline{I}) - 2I(I+1)J(J+1)}{2I(2I-1)2J(2J-1)} \quad (14)$$

$\underline{J}$ and $\underline{I}$ are the electronic and nuclear angular-momentum operator and a and b are the magnetic-dipole and the electric-quadrupole interaction constants. This leads to energy shifts due to magnetic dipole interaction

$$\Delta E_{md} = \frac{a}{2} [F(F+1) - J(J+1) - I(I+1)] \quad (15)$$

where F is the total angular momentum quantum number defined through the relation

$$\underline{F} = \underline{J} + \underline{I} \quad (16)$$

The energy shift due to the electric quadrupole interaction is given by

$$\Delta E_{eq} = b \frac{3K(K+1) - 2I(I+1) \cdot J(J+1)}{4I(I-1) \cdot 2J(2J-1)} \quad (17)$$

with

$$K = F(F+1) - J(J+1) - I(I+1) \quad (18)$$

The sixth term represents the isotope shift between atoms with different mass numbers A and A' corresponding to the masses M and M' . The isotope shift consists of two parts, the mass shift and the field shift. The mass shift arises from the fact that the nuclear mass is not infinite. The field shift is due to the finite volume of the nucleus which creates small differences in the potential energy. In the centre-of-mass system the kinetic energy can be expressed as

$$\frac{1}{2M} \underline{P}_n^2 = \frac{1}{2M} \left(-\sum_i \underline{P}_i \right)^2 = \frac{1}{2M} \sum_i \underline{P}_i^2 + \frac{1}{2M} \sum_{i \neq j} \underline{P}_i \cdot \underline{P}_j \quad (19)$$

where $\underline{P}_n$ is the momentum of the nucleus and $\underline{P}_i$ is the momentum of the i :th electron. The first term on the right-hand side gives rise to the normal mass shift and the second term causes the specific mass shift. The normal mass shift can be estimated by

$$\delta E_{NMS} \approx E \frac{m(M-M')}{MM'} \quad (20)$$

and the specific mass shift can be expressed in a similar way. The field shift can be written as

$$\delta E_{FS} = C \cdot \delta \langle r^2 \rangle^{AA'} \quad (21)$$

where C depends on the change in total electron density at the nucleus and $\delta \langle r^2 \rangle^{AA'}$ is the difference in mean-square radius between the two nuclei.

The seventh and eighth terms in the Hamiltonian correspond to the influence of magnetic and electric fields. Due to interaction between the magnetic moments of an atom and a weak external magnetic field B the energy levels are shifted an amount

$$\Delta E_m = \mu_B g_J M_J B \quad (22)$$

where μ_B is the Bohr magneton, g_J is the Landé factor and M_J is the magnetic quantum number. This so called Zeeman-effect causes a splitting into $2J+1$ sublevels.

The interaction between a homogeneous external electric field ϵ and an atom is called Stark effect and is given by

$$H_e = - \underline{\epsilon} \cdot \underline{D} \quad (23)$$

This operator has only non-zero matrix elements between states of different parity. A linear Stark effect can only be found in hydrogen and for highly excited Rydberg states. The lowest-order energy contribution to a state j is in other cases given by

$$\Delta E_e = \sum_i \frac{\langle j | \underline{\epsilon} \cdot \underline{D} | i \rangle \langle i | \underline{\epsilon} \cdot \underline{D} | j \rangle}{E_j - E_i} \quad (24)$$

If in a Rydberg series nearby levels, corresponding to $\Delta n=0$ and $\Delta l=\pm 1$, are taken into account the energy shift is according to Eq. 3 and Eq. 10

$$\Delta E_e \sim \frac{n^2 \cdot n^2}{n^{-3}} = n^7 \quad (25)$$

The energy-level shift can also be written as

$$\Delta E_e = -\frac{1}{2} \epsilon^2 [\alpha_0 + \alpha_2 \frac{3M_J^2 - J(J+1)}{J(2J-1)}] \quad (26)$$

where α_0 and α_2 are the scalar and tensor polarizabilities, respectively. An applied electric field will then cause both an energy level shift and a splitting corresponding to the different M_J -values.

II. NATURAL RADIATIVE LIFETIMES

II.1 Radiative Lifetimes and Oscillator Strengths

As a consequence of the Heisenberg uncertainty principle the energy width ΔE of an excited state with a lifetime τ is given by

$$\Delta E = \frac{h}{2\pi\tau} \quad (27)$$

This expression will normally set a resolution limit in all spectroscopic measurements. However, the resolution is improved if only long-lived atoms are detected [4].

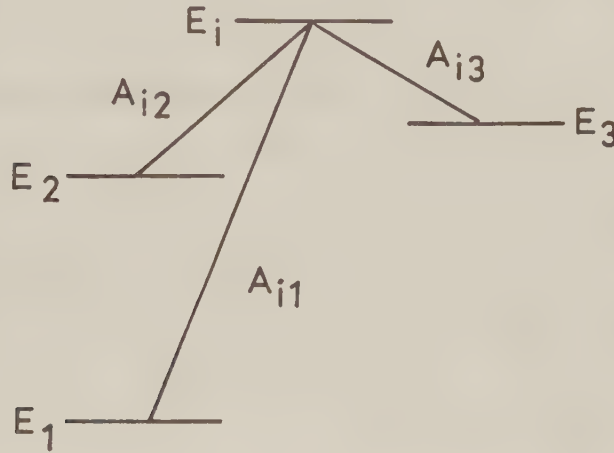


Fig. 1. Transition probabilities from an excited state E_i .

Let us consider an excited state E_i that has allowed electric dipole transitions to a number of lower lying states. The spontaneous transition probability per unit time to a lower state E_j is called A_{ij} . (See Fig. 1.) If the population N_i at level E_i is decreased with an amount dN_i during a small time interval dt the following relation holds

$$dN_i = -A_i N_i dt \quad (28)$$

with

$$A_i = \sum_j A_{ij} \quad (29)$$

Integration yields

$$N_i(t) = N_i(0) e^{-A_i t} \quad (30)$$

The light intensity I will then vary as

$$I(t) \sim \frac{-dN_i(t)}{dt} = A_i N_i(0) e^{-A_i t} \quad (31)$$

$$I(t) = I(0) e^{-A_i t} \quad (32)$$

The mean lifetime $\langle t \rangle$ of the excited state E_i can be calculated

$$\langle t \rangle = \int_0^{\infty} t e^{-A_i t} dt = \frac{1}{A_i} \quad (33)$$

which defines the lifetime τ_i of the state E_i as

$$\tau_i = \frac{1}{\sum_j A_{ij}} \quad (34)$$

The individual A_{ij} quantities can be expressed by using the electric-dipole operator $\underline{D}$.

$$A_{ij} = \frac{32\pi^3 \alpha c}{3\lambda_{ij}^3} |\langle i | \underline{D} | j \rangle|^2 \quad (35)$$

with

$$\underline{D} = e \sum \underline{r}_k \quad (36)$$

where $\underline{r}_k$ are the electron coordinates, α is the fine structure constant and λ_{ij} are the wavelengths of the transitions. As previously mentioned (Eq. 8), the matrix element squared is proportional to n^{-3} in a hydrogen atom. The lifetime values in a Rydberg series (1 constant) are then expected to scale as n^{*3} . Deviations from this rule can often be explained by the admixture of states with a different lifetime into the sequence. Lifetime values can then serve as a test of calculated wavefunctions or matrix elements and in some cases the admixture coefficients can be determined from lifetime data.

The oscillator strength f_{ij} of a transition is related to the transition probability per unit time A_{ij} by

$$f_{ji} = \frac{m \lambda_{ij} g_i}{4\pi\alpha h g_j} A_{ij} \quad (37)$$

where m is the electron mass and g_i and g_j are the degeneracy of the two levels. Oscillator strengths are required to do astrophysical calculations of element abundancies and temperatures.

II.2 Measurements of Radiative Lifetimes Using Laser Techniques.

Since reliable lifetime values are (apart from the pure atomic physics interest) essential to astrophysics, laser physics and plasma physics, a number of measuring techniques have been developed throughout the years [5,6]. In transitions that are connected with the ground state or a metastable state the natural line width can be used to determine the lifetime. In absence of Doppler broadening and collisions the line width determines the lifetime of the excited state (Eq. 27). With techniques that use radio-frequency transitions between sublevels, the Doppler broadening can be neglected and the lifetime is given by $\tau = 1/\pi\Delta\nu$.

In level crossing, measurements of changes in polarization of the fluorescence light is observed as a function of a variable magnetic field. Coherently scattered photons, corresponding to crossings of magnetic sublevels, will be observed as a linearly polarized component if the fluorescence light is detected in the magnetic field direction. The width of the crossing signals can be used to calculate the lifetime of an excited state. The strong zero field signal (Hanle signal) has been used to determine lifetimes of many low-lying states in the principal series. The method is sensitive to collisions and coherence narrowing and an extrapolation to zero atomic density must be made.

The most straight-forward method of measuring the radiative relaxation time is to excite a population of the studied system, terminate the excitation abruptly, and observe its decay. In the first experiments of this kind the excitation was accomplished by

an electron beam [7]. Along with the development of fast recording systems the technique evolved and a number of measurements on neutral and singly ionized atoms have been performed. Since the excitation is non-selective, cascade population from higher-lying states will effect the accuracy.

In the beam-foil technique an accelerated beam of fast ions passes through a thin target foil. Depending on the kinetic energy excited states in several times ionized species can be studied. The intensity of the decay light from a studied transition is measured as a function of the distance from the foil. With a knowledge of the beam velocity the distance can easily be translated into time. With the beam-foil technique very short lifetimes can be measured. However, the accuracy is limited by cascade population from higher lying states. With the use of multi-exponential fits to the decay curves, average accuracies of about 10% are obtained [8]. Beam-foil studies have also been made on neutral atoms.

Lifetime measurements using tunable dye lasers offer several advantages. The selective excitation will eliminate the problem of cascade population from higher-lying states. Using a time-resolved detection the studied decay will consist of only one exponential. The high powers per frequency unit available from a laser enable the use of atomic beams with low densities. The risk of systematic errors due to influence of collisions or multiple scattering is then small and time-consuming extrapolation to zero atomic density is eliminated. The laser techniques have mainly been used to measure lifetimes in neutral atoms, but by combining fast ion beams with laser excitation an increasing amount of data for ions is appearing [8].

a) Laser Light Sources

Laser action in organic dyes was discovered in 1966 [9,10]. Since dye molecules in a solvent have broad fluorescence profiles it became possible to produce tunable laser light. With a wavelength selective element inside the laser resonator, feedback is obtained only in a narrow band. By stimulated emission that light

is amplified and most of the available energy is transferred into a small wavelength region. A variety of tunable dye laser arrangements has been constructed [11,12].

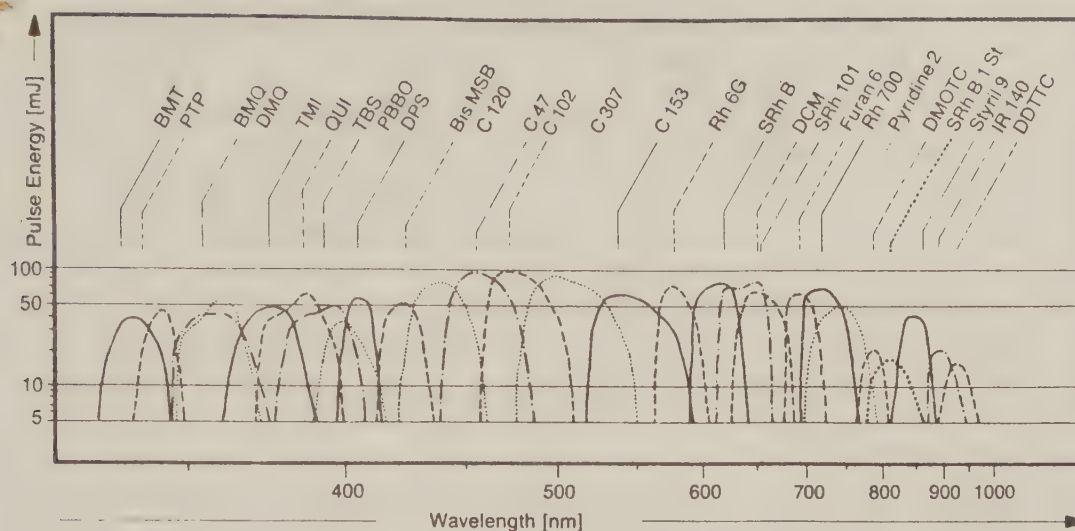


Fig. 2. Tuning curves for different laser dyes using excimer laser pumping. (From Lambda Physik FL 2002 dye-laser manual).

In order to get laser action from an organic dye, very intense optical pumping is required. This is achieved with flash lamps or fixed-frequency lasers. The most frequently used pump lasers are Nitrogen-, Nd:YAG- and Excimer lasers. They all produce short pulses (~ 10 ns) with very high peak powers (~ 1 -100 MW) but at a moderate repetition rate (~ 1 -100 Hz). The short duration of the pump pulses causes a fundamental Fourier limitation of the band width (~ 100 MHz) of the dye laser. In commercial dye lasers the band widths are of the order of 1-10 GHz. A number of laser dyes covers wavelengths from 320-1000 nm. (See Fig. 2.) Tunable laser light with pulse energies of the order of 10-100 mJ can be produced. The high peak powers enables frequency doubling and Raman shifting of the light and wavelengths down to 200 nm can be produced.

In order to obtain very narrow-band dye laser radiation, pumping from a continuous laser is required. An Argon-ion or a Krypton-ion laser then pumps a jet of dye solution inside a resonator [13]. The resonator can be designed for standing-wave or traveling-wave operation. Fig. 3 shows a drawing of a CW ring-dye laser. The wavelength is roughly selected by a Lyot filter and a

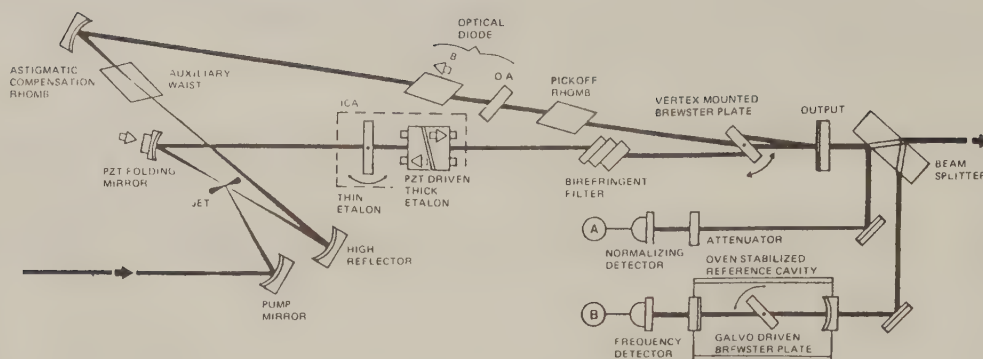


Fig. 3. Optical schematic of a single-mode ring dye laser.
(From Coherent Radiation CR-699 manual).

combination of two etalons favours only one longitudinal cavity mode. Using a so called optical diode, the light wave is forced to run in one direction only. If the resonator is stabilized by an electronic feed-back system the laser can produce CW light with a bandwidth of about 1 MHz. The feed-back system can also be used to sweep the frequency of the laser about 30 GHz. Output powers from a CW dye laser is of the order of 0.1 - 1 W in a single mode. With different laser dyes wavelengths in the visible and near IR region can be produced. By using frequency doubling and mixing in external cavities shorter wavelengths can also be produced [14].

The development of new laser dyes as well as other laser materials is rapid. Successful experiments with solid state materials has for instance lead to the construction of tunable F-center lasers [15, 16].

b) Pulsed Laser Excitation

With the use of dye lasers pumped by pulsed lasers high peak powers are obtained. Nonlinear processes can then efficiently be used to extend the available wavelength region. Two-photon absorption can also be used. Highly excited Rydberg states can be reached directly from the ground state or, for series with the same parity, in two-step excitation or two-photon absorption. (See Fig. 4.)

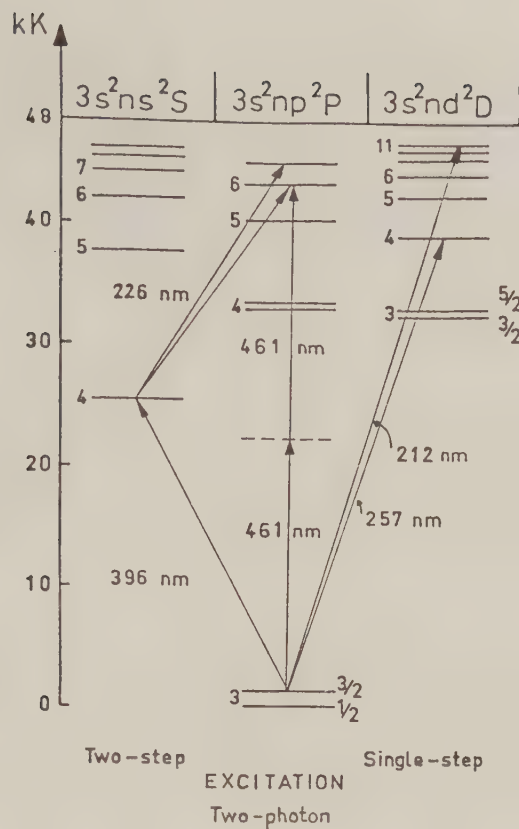


Fig. 4. Excitation scheme in Al showing two-photon absorption, single- and two-step excitation.

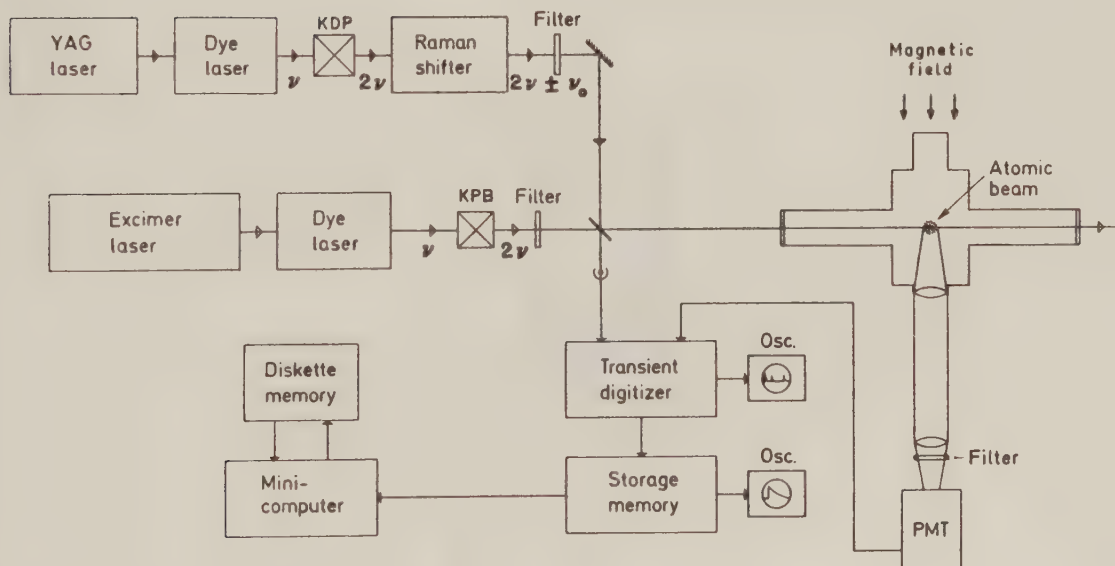


Fig. 5. Set-up used in the pulsed laser experiments.

Fig. 5 shows an experimental set-up used in our pulsed experiments. Notice that two separate light-producing systems were used. In some of the experiments a Nd:YAG laser was employed. The frequency doubled green light at 532 nm pumped a dye laser operating on green, yellow or red laser dyes. The tunable light was frequency doubled in a KDP-crystal and sent through a Raman shifter with high pressure H_2 gas. Each Stokes or anti-Stokes shift corresponds to a change of the photon energy by 4155 cm^{-1} ($\sim 0.5\text{ eV}$). In an anti-Stokes shift the photon energy is increased. With a suitable combination of laser dye, doubling crystal and a number of anti-Stokes shifts, wavelengths down to 200 nm can be produced.

In most of the pulsed experiments an excimer laser operating on XeCl gas mixture was used. The 308 nm light pumped a tunable dye laser usually operating on blue dyes. In the case of two-step excitation part of the excimer light was split off to pump a second homebuilt Littman-cavity dye laser [12]. The produced dye laser light could be frequency doubled with the use of a KDP- or a KPB-crystal. Tunable light down to 217 nm could be used in one- or two-step excitation schemes.

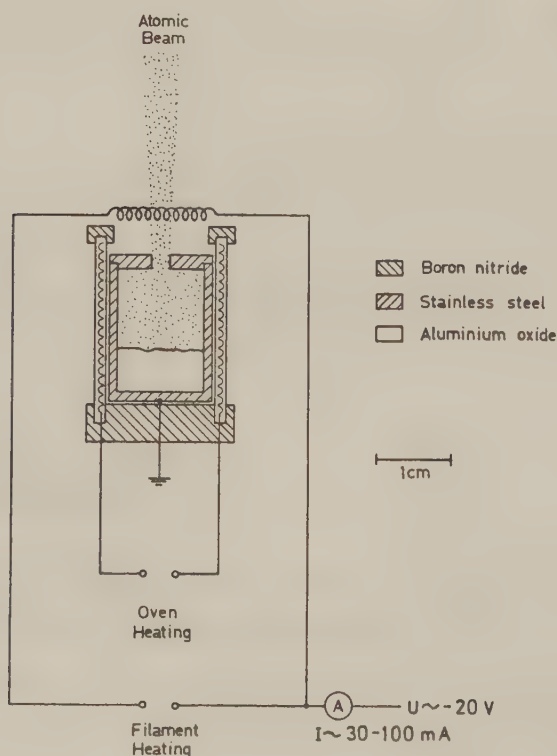


Fig. 6. An atomic-beam oven design.

The atomic beam was created in a vacuum system using a resistively heated oven. (See Fig. 6.) Through a discharge in the atomic beam metastable atoms could be produced. The atomic beam was terminated on a liquid nitrogen cool trap. The magnetic field in the excitation volume was controlled by two sets of Helmholtz coil systems.

The light released in the decay passed through appropriate interference filters and coloured glass filters and was detected with a photomultiplier tube (PMT). The electric signal was digitally converted in a fast transient recorder and added in a storage memory. When an appropriate signal-to-noise ratio had been reached the decay curves were stored on a floppy-disc. Using Newton's method, a nonlinear least-squares fit to an exponential was performed in a minicomputer. Fig. 7 shows the quality of three experimental curves obtained with the pulsed laser technique.

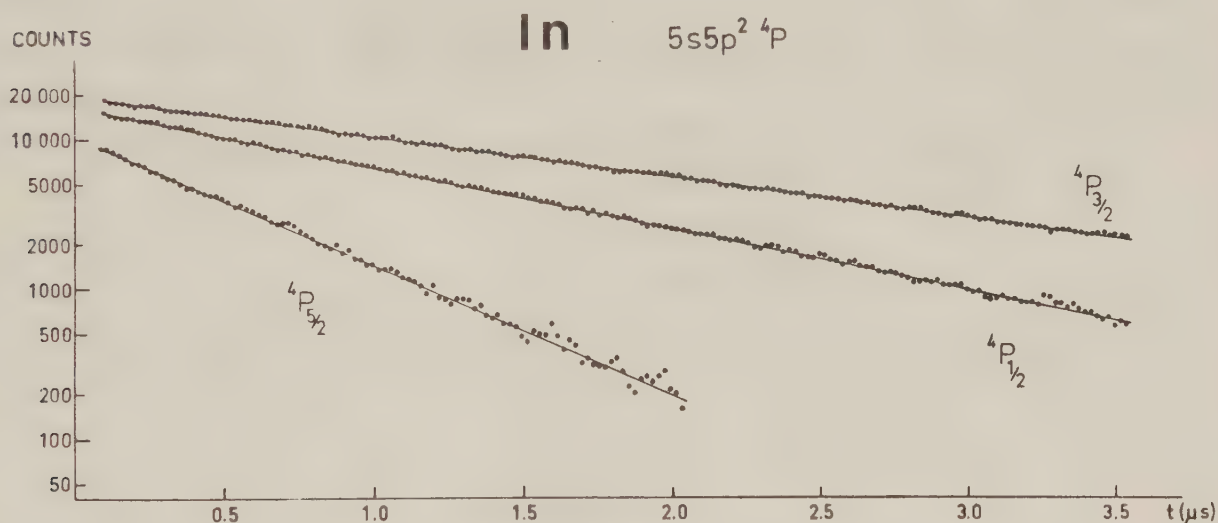


Fig. 7. Three experimental decay curves from the $5s5p^2 4P$ states in In, shown on a logarithmic scale. Notice that the lifetimes are J-dependent.

c) CW Laser Excitation

Modulated CW laser light provides some interesting features. The bandwidth of a CW laser can be as small as 1 MHz. This enables

the distinction of very close-lying atomic levels. For instance it may be possible to measure lifetimes on different isotopes of an element. Continuous light may be modulated at a very high frequency ($\sim$ MHz), which makes the delayed-coincidence method favourable. This technique is particularly free from systematic effects due to nonlinearities etc.

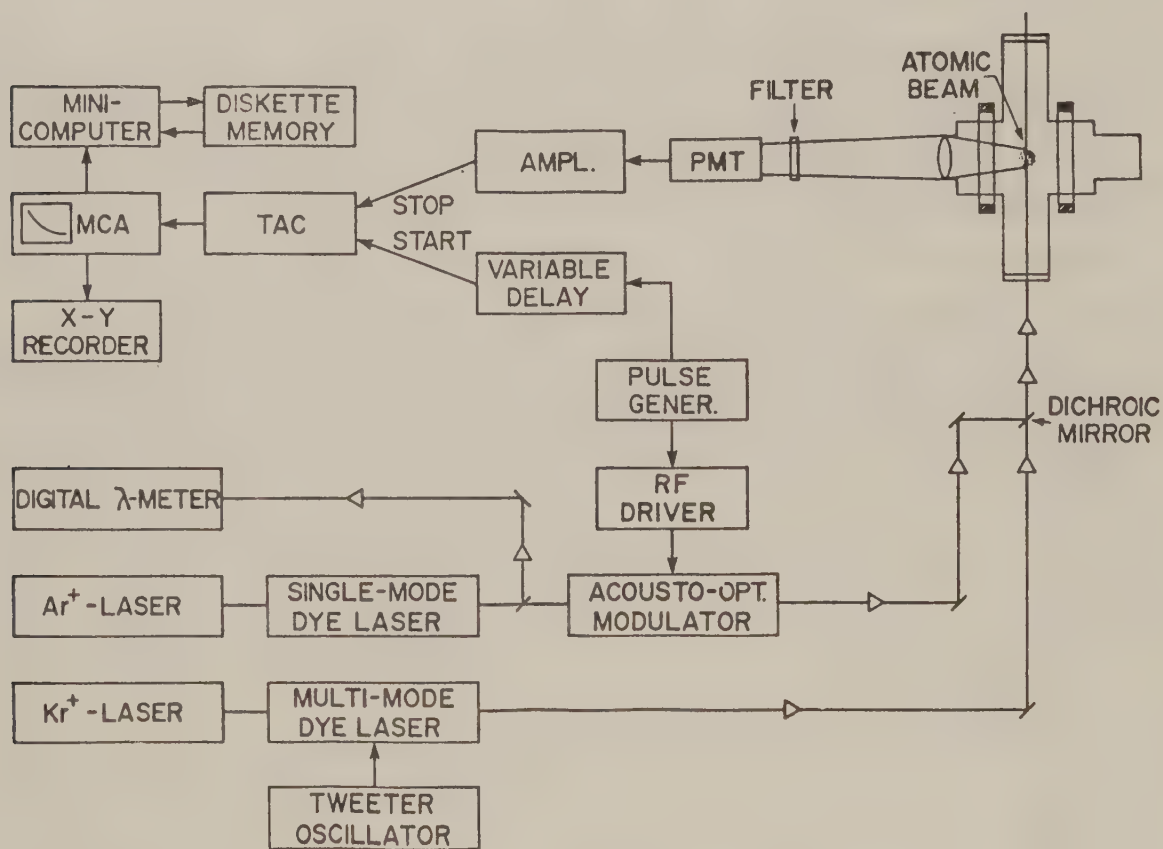


Fig. 8. Experimental set-up used in the delayed-coincidence measurements.

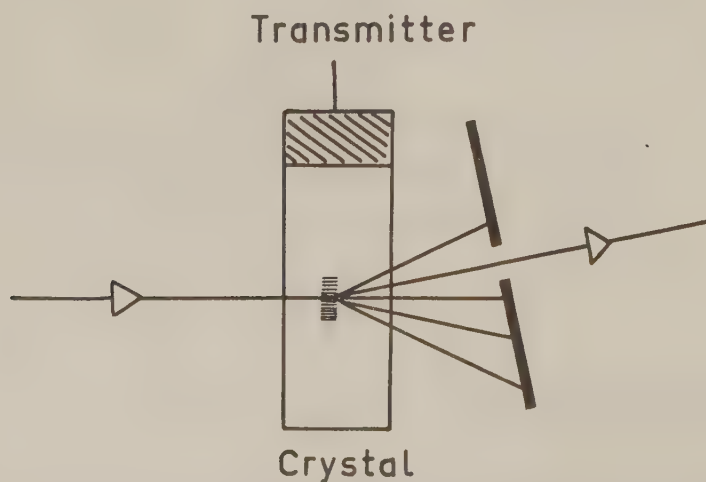


Fig. 9. A schematic drawing of an acousto-optic modulator.

Fig. 8 shows the experimental set-up in a PUMOLS-experiment (Pulse MODulated Laser Spectroscopy). The atomic-beam apparatus is similar to the one used in the pulsed experiments. Light from a tunable dye laser (in step-wise excitation the second-step laser) is sent through an acousto-optic modulator. In the modulator the light is focused in a transparent crystal and exposed to high-frequency acoustic waves. The acoustic waves change the refractive index of the crystal and the light sees a transmission grating passing by. (See Fig. 9.) In absence of acoustic waves the light is going straight through the crystal. With an aperture, the first-order diffraction beam can be isolated and used in the experiment. With a pulse generator coupled to the acousto-optic modulator, light pulses with suitable length and duty cycle can be produced. The pulse generator also delivers (after a variable time delay) a start pulse to the time-to-amplitude converter (TAC). The first photon that reaches the PMT is used as a stop signal for the TAC. The used procedure is explained in Fig. 10. The created pulses with different amplitudes are collected in a multi-channel analyser (MCA). The decay curves were transferred to a minicomputer and stored on a floppy-disc. Fig. 11 shows the quality of a PUMOLS-recording.

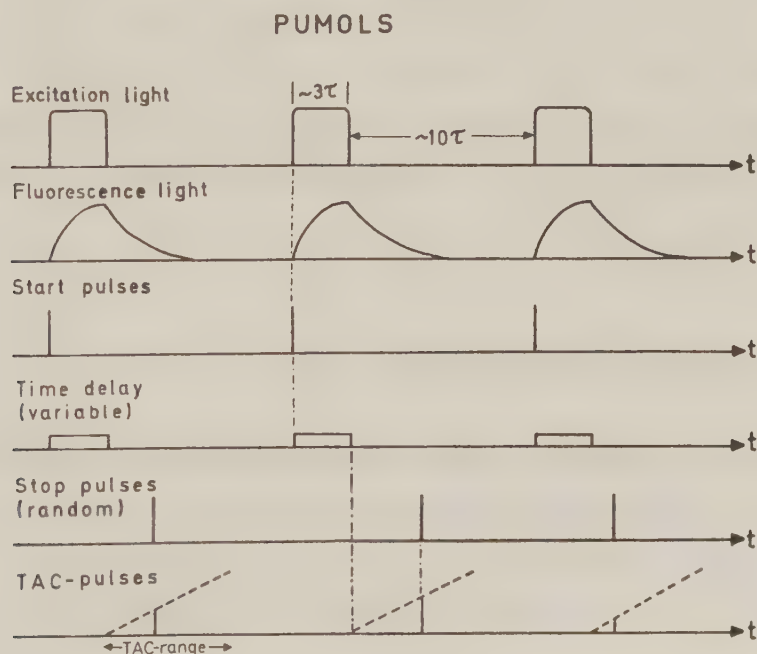


Fig. 10. Pulse scheme in a PUMOLS experiment.

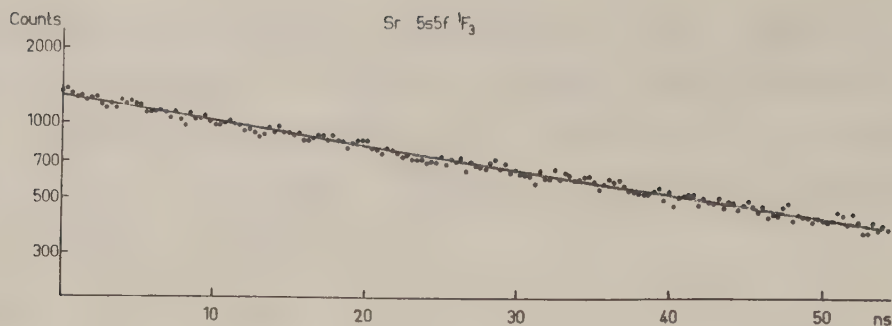


Fig. 11. PUMOLS recording of the $5s5f\ ^1F_3$ state in Sr.
The decay curve is shown on a logarithmic scale.

To avoid dead time in the TAC we sometimes interchanged the start- and stop signals [17]. The TAC is then activated only when a photon arrives at the PMT.

Since only the first fluorescence photon is counted in delayed-coincidence experiments it is important that the probability of a second photon arriving at the PMT within a duty cycle is very small. Otherwise the method will produce a pile-up effect, i.e. the amplitudes corresponding to short times will be overrepresented. (The result would be a too short lifetime value.) Experimentally one detected photon on thirty duty cycles will produce a completely negligible pile-up. In some detection systems pile-up can be electronically rejected [18]. Normally, the delayed-coincidence method excludes the use of pulsed lasers. With pulse frequencies around 10 Hz the sampling time to obtain a useful decay curve is very long.

d) Evaluation of Decay Curves

When the lifetime of a studied state is long compared to the risetime (or rather the decay time) of the excitation pulse it is easy to calculate the lifetime value. Apart from the background the fluorescence decay will then best be described by an exponential.

If a lifetime is short compared to the decay time of the excitation pulse precautions must be taken to avoid systematic errors. Normally an exponential behaviour would be expected some time

after the excitation pulse is turned off. In practice the signal-to-noise ratio will not always allow the evaluation to start at a sufficiently late time. If the lifetime is very short the trailing edge of the excitation pulse must always be taken into account.

To evaluate short lifetimes the following model was used. A small part of the excitation pulse can be regarded as a square pulse excitation, which will cause an exponential decay. The excitation pulse can be recorded and stored as subsequent square pulse elements. For each element a computer can generate a decay curve and add up a theoretical fluorescence curve for a certain lifetime value. The computer can also perform fits to the experimental fluorescence curves using the lifetime as a parameter. This procedure will provide good fits in the absence of saturation and straylight. It is then favourable if the fluorescence is not detected on the laser wavelength.

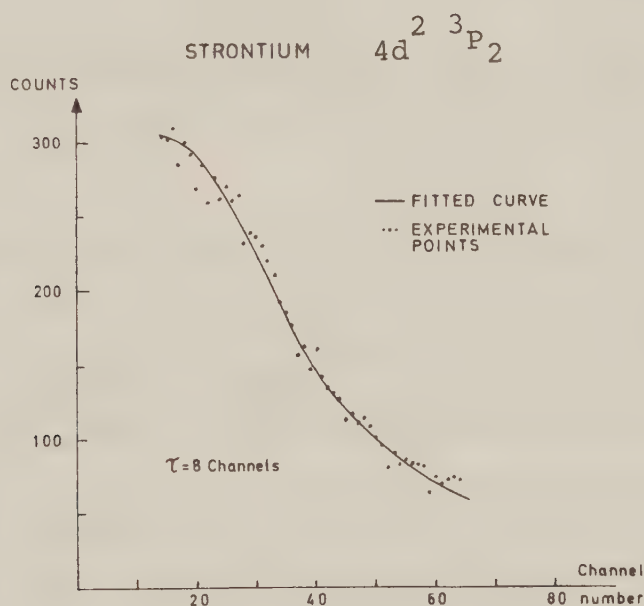


Fig. 12. PUMOLS recording of the short-lived $4d^2 \ ^3P_2$ state in Sr. The fluorescence light was recorded at an UV transition. The best fitted computer-generated curve is included in the figure.

Fig. 12 shows a fluorescence recording of a short-lived state in Sr obtained with the PUMOLS technique. The fitted computer generated curve is also included.

In order to better control the recordings of the laser pulse, a movable rod could be inserted into the excitation volume, without affecting the high vacuum. The rod was adjusted to create similar recording conditions for both laser pulse and fluorescence. Fig. 13 shows a fluorescence curve from a state in Al obtained with the transient digitizer. Included in the figure are also recordings of the excitation pulse and the best fitted computer generated fluorescence curve. In spite of excellent fits rather large error limits were set to cover possible systematic errors.

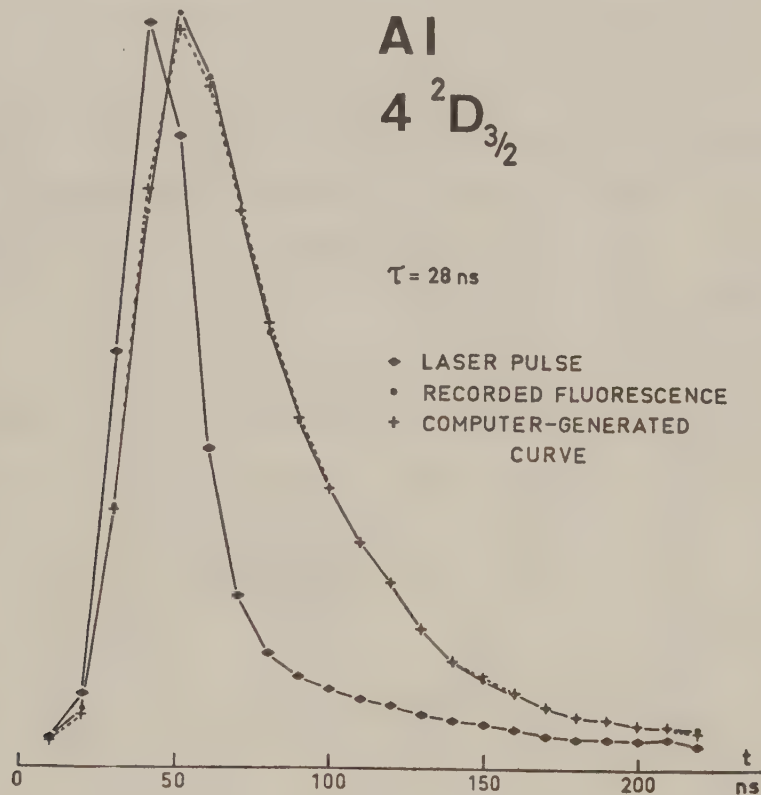


Fig. 13. A recording of the fluorescence light from the $4d \ ^2D_{3/2}$ state in Al. Included in the figure are also a recording of the excitation pulse and a computer-generated curve.

If the excitation light is modulated in a sinusoidal manner, with a frequency Ω , the fluorescence light is also expected to be sinusodially modulated with the same frequency but phase-shifted [5] by an amount φ according to

$$\tan \varphi = \tau \Omega \quad (38)$$

where τ is the lifetime of the excited state. (There is an analogy to an electric RC-circuit having a time constant τ .) Fig. 14 shows two experimental curves obtained with the PUMOLS version of the phase-shift technique. To obtain accurate values

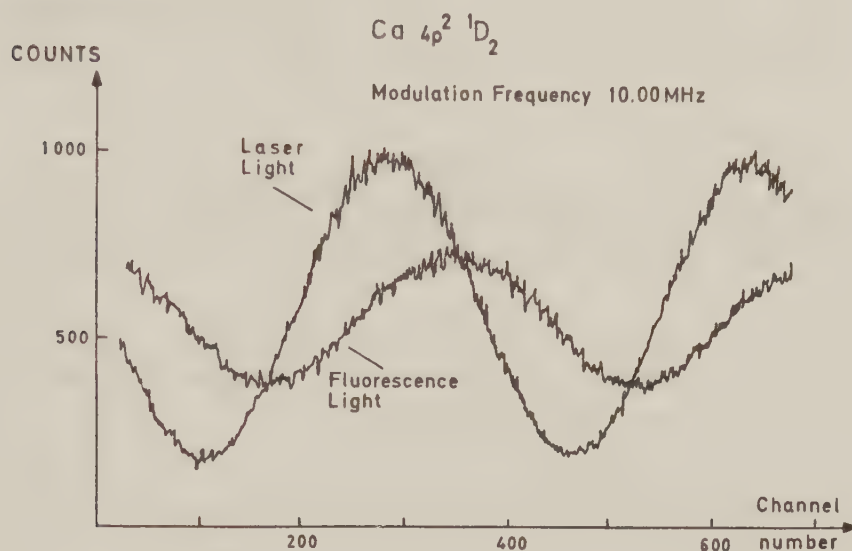


Fig. 14. Two experimental curves obtained with the PUMOLS version of the phase-shift technique.

of the phase-shift every set of curves had to be computer treated. The precision of the method is in our case mainly limited by the ability of the acousto-optic modulator to produce sinusoidal modulation. However, it is in principal possible to calculate lifetimes with the phase-shift technique even if the pulse shape is far from sinusoidal. Any excitation pulse can be expanded in a series of sinewaves with different amplitudes and frequencies. Each sinewave is then delayed a time t by the atoms

$$t = \frac{\arctan \Omega \tau}{\Omega} \quad (39)$$

Fig. 15 shows how square-wave excitation is gradually being transformed into the phase-shift scheme due to non-zero rise and decay time of the excitation pulse. It is also possible to say that the same figure shows the gradual transformation of the phase-shift method from a pure case to a superposition of several phase-shifted sinewaves.

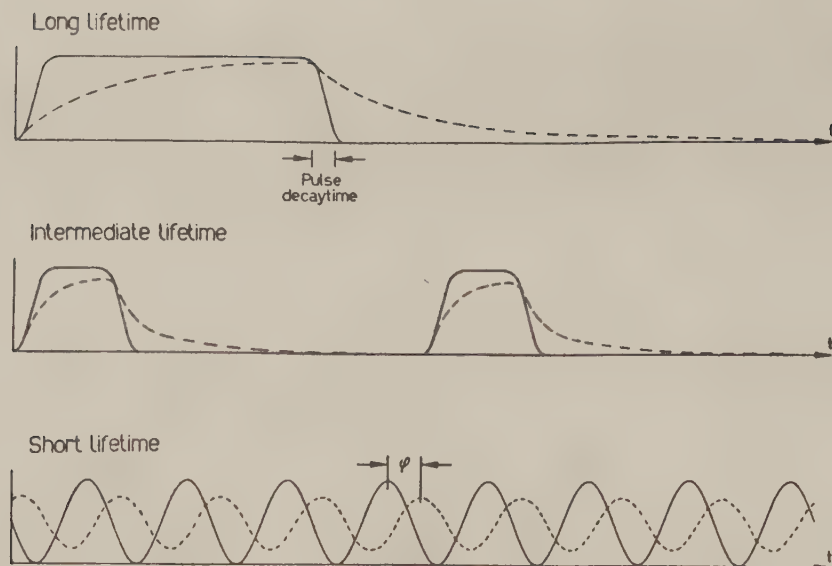


Fig. 15. The gradual transformation of square-wave excitation into the phase-shift scheme.

e) Sources of Error

Both short (< 20 ns) and long lifetimes (> 7 μ s) are difficult to measure with the described methods.

The accuracy in measurements of long lifetimes is mainly limited by blackbody-induced transitions. In a Rydberg series the number of levels per energy interval scale as n^3 . In a highly excited atom there are a number of levels that can be populated through interaction with the microwave background at room temperature. The studied atoms can both be excited to a higher level and deexcited through stimulated emission. The lifetime values in a Rydberg sequence with only blackbody transitions are expected to scale as n^2 [19]. The experimental lifetime τ_{exp} then consists of a lifetime for dipole transitions to low-lying states τ_{dp} and a blackbody contribution τ_{bb}

$$\frac{1}{\tau_{\text{exp}}} = \frac{1}{\tau_{\text{dp}}} + \frac{1}{\tau_{\text{bb}}} \quad (40)$$

The result will be an extra channel of depopulation and consequently the measured lifetime is decreased. Cases with $\tau_{\text{dp}}/\tau_{\text{exp}} \sim 2$ have been reported [20]. Experimental attempts

have been made to minimize the influence of blackbody induced transitions [21].

When it comes to very long lifetimes the imaging of the decay region onto the PMT can result in a flight-out-of-view effect. The mean kinetic energy $\langle W_k \rangle$ in the atomic beam is determined by the temperature T as

$$\langle W_k \rangle = 3kT \quad (41)$$

where k is the Boltzmann constant. The mean velocity $\langle v \rangle$ can then be approximately written as

$$\langle v \rangle \approx \sqrt{\frac{6kT}{M}} \quad (42)$$

where M is the atomic mass. $\langle v \rangle$ is of the order of millimeters per microsecond. This means that very long-lived atoms decay out of sight and the decay curve will be distorted.

The pressure in the atomic beam and in the vacuum system can affect the experimental lifetime. At high atomic densities the studied atoms may be deexcited through collision processes. That would lead to a shortening of the measured lifetime in long-lived states. Short-lived states that are connected with the ground state (or a metastable state) through a strong transition may be affected by multiple scattering (photon trapping). A released photon can then be absorbed and reemitted several times before it reaches the PMT. The result would be a longer observed lifetime value.

Another possible source of error is due to quantum beats arising from the Zeeman splitting in a weak magnetic field. (The quantum beat phenomenon is described in the next chapter). If the period of the beats is long, corresponding to small splittings, the modulation may destroy the exponential decay curve and introduce a systematic error. It can not be excluded that the small magnetic field due to the earth may cause such an effect. However, if the period of the quantum beats is very short, corresponding to large splittings, the beats can not be resolved by the detection system. During the measurements a sufficiently large magnetic

field was applied in the excitation volume to avoid influence of slow Zeeman quantum beats.

f) Computer Simulations

In order to obtain information on the interaction between the detection system and the used computer routines several simulations of the experiments have been made.

The simulations were all based on an algorithm that produced exponentially distributed pseudo-random numbers. If every produced random number was counted within a set of 1024 windows this would correspond to the PUMOLS experiments. When a decay curve was generated the lifetime and the number of detected pulses was set. Before estimating the lifetime noise was added to the decay curve. With the use of random noise the computer programs always made a too long estimate of the lifetime when the signal-to-noise ratio was bad. This is supported by experimental experience. When sinusoidal noise was added, corresponding to detected Zeeman quantum beats, the lifetime programs made good estimates of the lifetimes as long as the period of the sine-waves was shorter than the lifetime. It is also possible to create decay curves with a pile-up effect. It was then easy to control that the experimentally used ratios between start and stop pulses gave a negligible pile-up effect.

In order to simulate the pulsed experiments every random number was ascribed a (variable) distribution curve. By choosing different curve shapes with different heights various aspects of the detection system could be analysed. If every random number is given a rectangular distribution this will correspond to box-car detection. If every exponential random number was assigned for instance a Lorentzian pulse shape with a random height in 256 steps an attempt to simulate transient digitizer detection was made. This would then corresponds to single photons arriving at the PMT. It should be noticed that the detection of a distribution in itself introduces noise compared to the delayed coincidence technique. It is also possible to simulate saturation

effects in the PMT, corresponding to many detected photons after every laser pulse.

The aim of the computer simulation was to understand the detection system in combination with our computer routines and a great deal of confidence and insight in our experiments was achieved during the many test runs.

III. HYPERFINE STRUCTURE MEASUREMENTS

III.1 Quantum-Beat Spectroscopy

If two (or more) close-lying atomic levels are simultaneously excited by a short laser pulse, the fluorescence decay is modulated by a frequency (or by frequencies) which is (are) related to the energy separation(s) in the excited state. This phenomenon, known as quantum beats, is a quantum mechanical interference process that can be compared with an optical double-slit experiment. The process is fully explained using quantum electrodynamics [22], but the basic principle can be understood with a semi-classical approach [23].

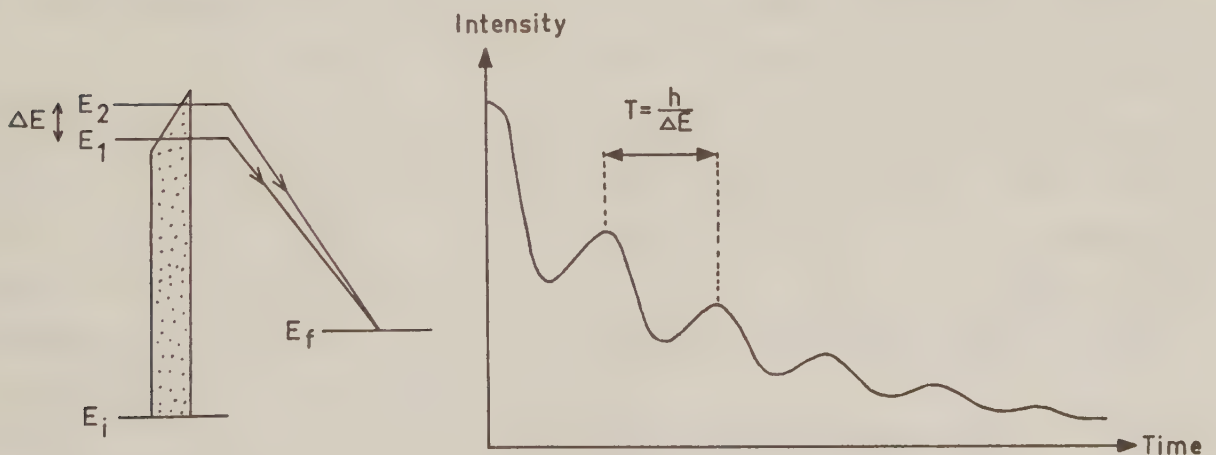


Fig. 16. Excitation and detection in a quantum-beat measurement.

Two closely spaced levels E_1 and E_2 are pumped by a short laser pulse with a spectral width larger than the energy separation ΔE . (See Fig. 16.) The wavefunction of the excited state can then be written as a superposition of the wavefunctions of the two states

$$\Psi = a_1 \varphi_1 + a_2 \varphi_2 \quad (43)$$

where a_1^2 and a_2^2 are the probabilities of finding the atom in state E_1 and E_2 , respectively. Now if the decay to a final state E_f is considered, the excited state can classically be written as a sum of two damped oscillators in the following way

$$\Psi(t) = a_1 \varphi_1 e^{-i\omega_1 t - \frac{t}{2\tau_1}} + a_2 \varphi_2 e^{-i\omega_2 t - \frac{t}{2\tau_2}} \quad (44)$$

where

$$\omega_i = (E_i - E_f)/\hbar, \quad i = 1, 2 \quad (45)$$

and τ_i are the lifetimes of the excited state. The time-dependent intensity of the decay light is then given by

$$I(t) = \text{constant} \cdot |\langle \varphi_f | \underline{D} | \Psi(t) \rangle|^2 \quad (46)$$

where φ_f is the wavefunction of the final state f and $\underline{D}$ is the dipole operator. Assuming that $\tau_1 = \tau_2 = \tau$, (which is the case for sublevels of a given atomic state) we have

$$I(t) = \text{constant} \cdot e^{-\frac{t}{\tau}} (A + B \cos \omega_{21} t) \quad (47)$$

with

$$A = a_1^2 |\langle f | 1 \rangle|^2 + a_2^2 |\langle f | 2 \rangle|^2 \quad (48)$$

$$B = a_1 a_2 |\langle f | 1 \rangle| |\langle f | 2 \rangle| \quad (49)$$

$$\omega_{21} = \omega_2 - \omega_1 \quad (50)$$

Thus if both matrix elements are non-zero, the decay light is modulated on a frequency corresponding to the energy separation between E_2 and E_1 . Measurements of the modulation frequency ω_{12} allows a determination of energy splittings that would be obscured by Doppler broadening. The resolution of the quantum-beat method is only limited by the natural linewidth of the excited states.

As a result of the quantum theory of measurements all attempts to distinguish between the two decay channels will destroy the beats. Let us for instance say that the decay light from level E_1 can, due to selection rules, be excluded by the use of polarizers. In that case the matrix element $\langle f|1\rangle$ is zero and no beats will appear. Another way of distinguish between the decay channels would be to use a very narrow-band filter centered at one of the frequencies ω_1 or ω_2 . If the filter is centered around ω_1 the amplitude of ω_2 will vanish and again the beats disappear. From a quantum mechanical point of view the two states (1 and 2) are said to be indistinguishable. This is not the case for an energy-splitting in the lower state. If the lower state consists of two levels E_f and E_f , there is (in principle) nothing that prevents an investigation of which state the atom ended up in long after the decay from E_1 or E_2 . Thus the corresponding amplitude can not interfere and an energy splitting in the lower state will not cause beats in a single atom. In a many-atom system beats arising from a lower level splitting is theoretically possible [22]. It would however require a high atomic density and the signal would no longer be Doppler-free.

So far little has been said about the origin of the energy splitting. It could for instance be a fine structure or a hyperfine structure splitting. The energy level splitting could also be caused by electric or magnetic fields. In our case only hyperfine structure has been studied with the quantum-beat technique. The first hfs investigation using quantum-beat spectroscopy with lasers was made in 1973 [24]. Along with the development of fast transient recorders the quantum-beat method has become a valuable spectroscopic tool.

The experimental set-up used in our quantum-beam experiments is the same as the one used in the pulsed lifetime measurements (See

Fig. 4) but two linear polarizers are added, one in the laser beam and the other on the detection side. (The laser beam is originally only partly linearly polarized.) To be able to set the angle θ between the two polarizers a prism polarization rotator was also inserted in the laser beam. The modulation amplitude of the beats is proportional to $(3 \cos^2 \theta - 1)$ [23]. This means that with $\theta = 54.7^\circ$ the beats will vanish and only an exponential is detected. If the polarizer setting is changed from perpendicular to parallel the beat phase is changed by 180° and the beat amplitude is doubled. If two such curves with opposite phase are subtracted the exponential curve is suppressed and the beat structure (exponentially damped) is enhanced.

The selection rules for electric dipole transitions in the case of hyperfine structure are $\Delta F = 0, \pm 1$. This gives beat frequencies corresponding to $\Delta F = \pm 1, \pm 2$. If for a moment the electric-quadrupole interaction is neglected the beat frequencies can easily be expressed in the magnetic-dipole constant a using the

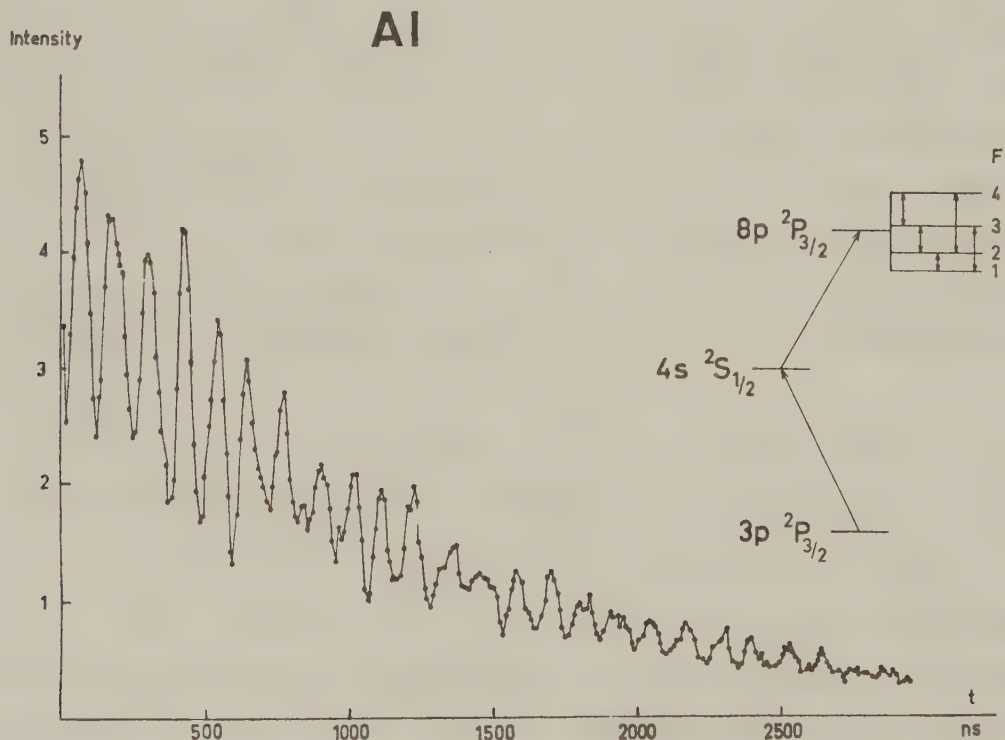


Fig. 17. Quantum beats in the $8p \ ^2P_{3/2}$ state in Al. Energy differences that can cause beats are indicated in the figure.

Landé interval rule. If the studied structure has more than two levels several beat frequencies will occur in the fluorescence decay. (See Fig. 17.) The best way to determine the different beat components is to do a Fourier transform of the time-resolved recording.

The Fourier cosine transform of the simple time spectrum in Eq. 47 is

$$I(\omega) \sim \frac{A \cdot \frac{1}{\tau}}{\omega^2 + (\frac{1}{\tau})^2} + \frac{\frac{1}{2}B \frac{1}{\tau}}{(\omega - \omega_{12})^2 + (\frac{1}{\tau})^2} + \frac{\frac{1}{2}B \cdot \frac{1}{\tau}}{(\omega + \omega_{12})^2 + (\frac{1}{\tau})^2} \quad (51)$$

With ω and $\omega_{12} > 0$ the frequency spectrum exhibits one peak at $\omega = 0$ and another at $\omega_{12} \approx 0$ (The position may be slightly moved due to overlap.) The peaks have a full width at half maximum of $2/\tau$ and are separated only if $\omega_{12} > 1/\tau$. (The natural radiation width corresponding to the Heisenberg uncertainty relation.)

At the same time as the Fourier transform is applied a biasing function $f(t)$ can be introduced

$$G(\omega) = \int_0^{\infty} I(\omega) f(t) \cos \omega t dt \quad (52)$$

In an experiment the evaluation often starts after some time t_1 corresponding to a bias function

$$f(t) = \begin{cases} 0 & t < t_1 \\ 1 & t > t_1 \end{cases} \quad (53)$$

This form of bias introduces an oscillatory structure (with a period of $1/t_1$) into the wings of the spectral peaks.

If the time spectrum is biased with an increasing exponential

$$f(t) = K e^{t/q} \quad q > \tau \quad (54)$$

the Lorentzian shape is preserved but the peaks are narrowed from $2/\tau$ to $2/\tau - 2/q$. In an experimental case this must be combined with a cut-off after some time and again oscillatory structure is introduced.

A number of different bias functions can be used to narrow the lines and suppress the spurious peaks. (The bias functions are sometimes called apodisation functions since they are taking away the foot of the frequency peak.) A narrowing of the peaks always results in a decrease in the signal-to-noise ratio and a balance must be made between attempts to resolve close-lying lines and the possibility of destroying weak frequency components.

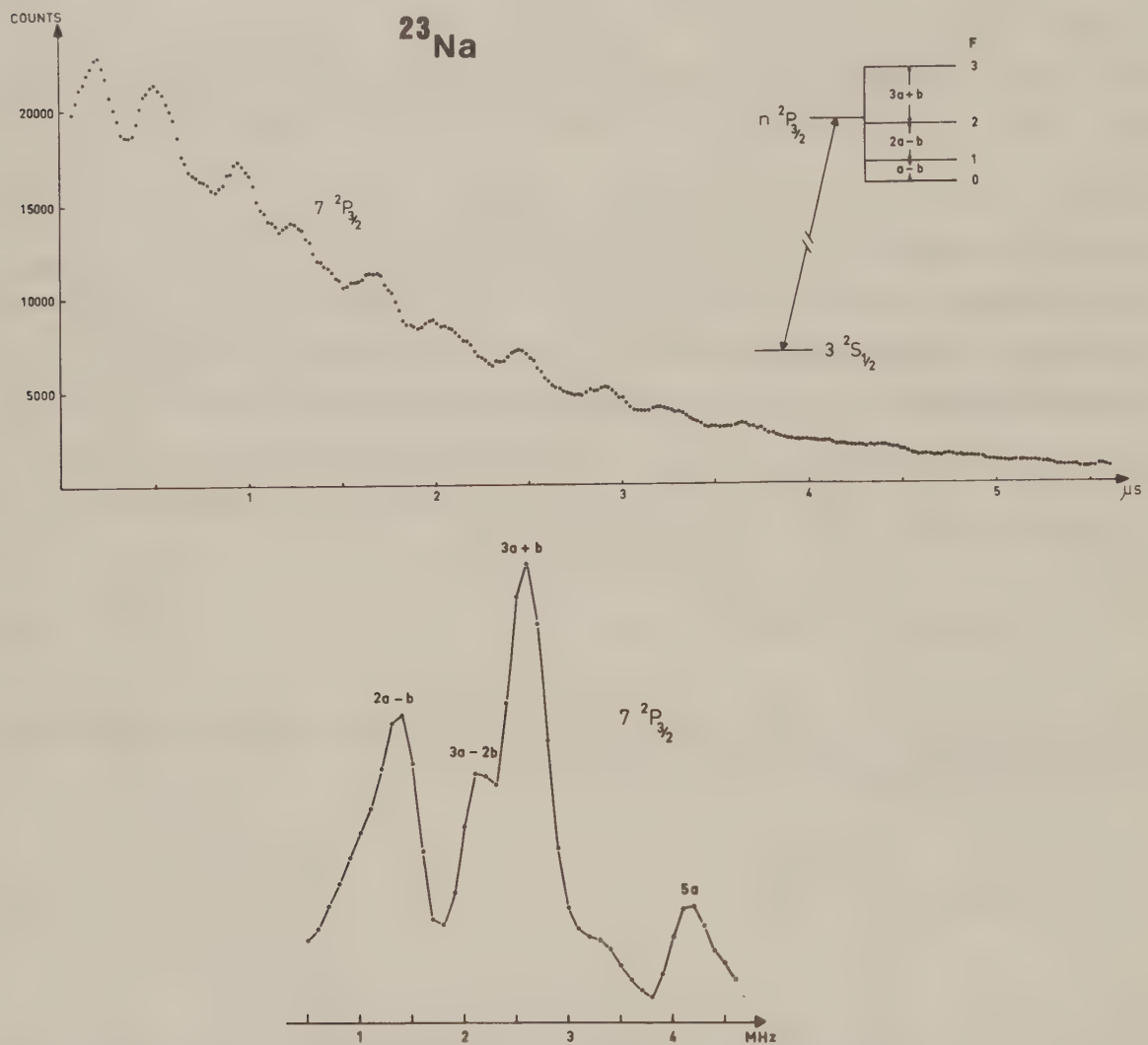


Fig. 18. A quantum-beat recording of the $7\ ^2P_{3/2}$ state in ^{23}Na and the corresponding frequency spectrum obtained using a cosine-square apodisation function.

Fig. 18 shows an experimental quantum-beat recording of the $7\ ^2P_{3/2}$ state in ^{23}Na and the corresponding frequency spectrum. It should be noticed that in the absence of electric-quadrupole interaction the peaks corresponding to $F=3 \leftrightarrow F=2$ and $F=2 \leftrightarrow F=0$ should coincide.

During the measurements the earth magnetic field in the excitation volume was compensated for. This was obtained by adjusting away Zeeman-quantum beats in the $6s6p\ ^3P_1 - 6s^2\ ^1S_0$ transition in Yb. (A magnetic field of 10^{-4} T will here cause beats with a frequency of about 4.4 MHz.)

In a quantum-beat measurement only energy level splittings are determined. The relative energy positions between different isotopes can not be seen since quantum beat is a phenomenon taking place in one atom at a time. This also means that the frequency spectrum from species with e.g. more than one hyperfine structure may be difficult to analyze if the peaks are overlapping. Since energy-level splittings are deduced with the quantum beat method, only the relative signs of the hyperfine structure constants can be determined. The absolute values must be determined by other means.

As previously mentioned, beats corresponding to small splittings (low frequencies) can not be measured if the lifetime is short. The largest splittings that can be measured in fluorescence detection is determined by the fast transient digitizer and the PMT-response. The present upper limit should be somewhere around 200 MHz. With the use of so called fast beam-laser technique splittings up to 10 GHz can be determined [25].

III.2 High-Resolution Laser Spectroscopy on a Collimated Atomic Beam

With the use of tunable narrow-band dye lasers, high-resolution measurements can be performed with a number of techniques such as saturation spectroscopy, polarization spectroscopy and Doppler-free two-photon absorption [5,26,27]. All these techniques can reveal structures that normally are concealed by Doppler broadening. The frequency width due to Doppler broadening in a transition is given by

$$\Delta\nu_D = 7.16 \cdot 10^{-7} \nu \sqrt{\frac{T}{A}} \quad (55)$$

where T is the temperature and A is the mole weight. In a cell $\Delta\nu_D$ is of the order of 1 GHz which is large enough to obscure for

instance hyperfine structures in highly excited states. In an atomic beam the velocity components along the laser beam and in the detector direction can be reduced with apertures. In a highly collimated atomic beam the spectral width of a signal is mainly determined by the natural line width. In two-step excitation schemes it is normally a short-lived intermediate state that sets the resolution limit.

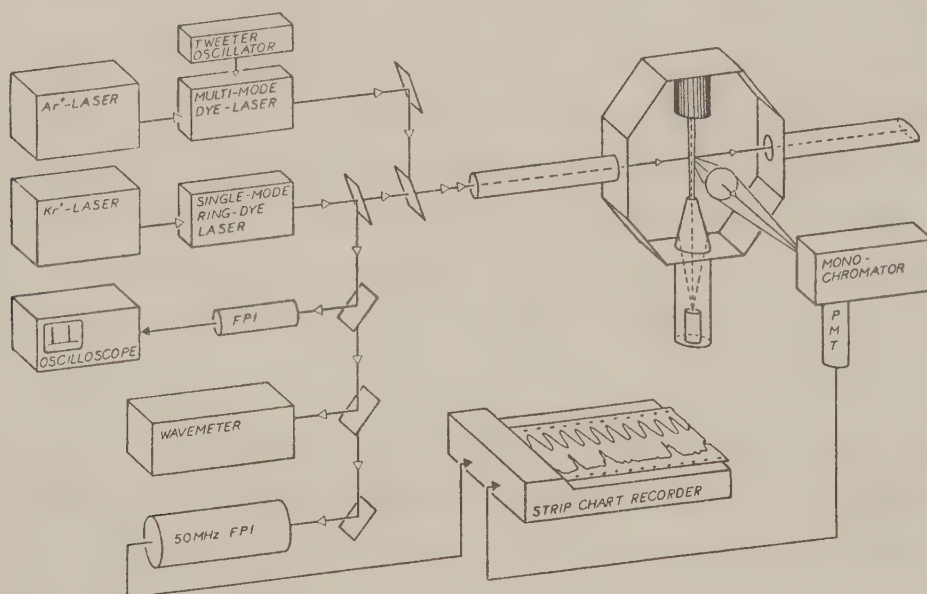


Fig. 19. Experimental set-up used in the high-resolution measurements on a collimated atomic beam.

Fig. 19 shows an experimental set-up used in high-resolution measurements on a collimated atomic beam. The laser beam and the detector are at right angles to the atomic beam. Part of the (second-step) laser light is split off and sent to a 50 MHz multi-pass interferometer to provide a frequency scale when the laser is scanning. Fig. 20 shows an experimental recording of a transition in Ba. From such recordings hyperfine structure constants and isotope shifts can be deduced. With the same type of experimental set-up it is also possible to study the influence of electric and magnetic fields on atomic transitions.

In quantum-beat spectroscopy the laser light is turned off during the signal recording. Power broadening and shifts caused by the laser light can then not affect the result. In the high-resolution measurements precautions must be taken to ensure that the intense laser light is not giving rise to systematic errors.

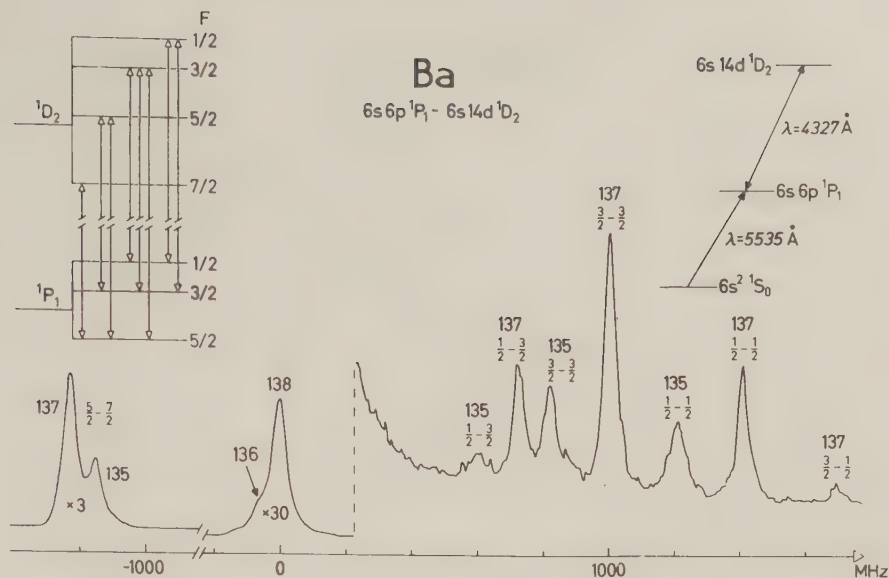


Fig. 20. Experimental recording on a hfs in Ba obtained with high-resolution spectroscopy on a collimated atomic beam.

IV. COMMENTS ON THE RESULTS

IV.1 Radiative Lifetimes

The radiative lifetimes in an unperturbed Rydberg series are expected to scale as $(n^*)^k$, where k is close to three. For instance this is the case in the measured S sequences in Mg, In and Al. (See Fig. 21.) Since these sequences are unperturbed, theory should be able to predict the lifetime values with good accuracy. In the S-sequences in In and Al calculated values based on the Coulomb approximation [28] are in agreement with the experimental data in In, but in Al all the calculated values are about 30% lower. Some lifetime values in the S sequence in Mg were compared with results obtained from multi-configurational Hartree-Fock calculations [29] and an excellent agreement was found.

If a sequence is perturbed by the admixture of other states, the lifetime values would deviate from the simple scaling rule. In Ba it has been shown that the admixture of a short-lived doubly excited state can cause a large decrease in an expected lifetime

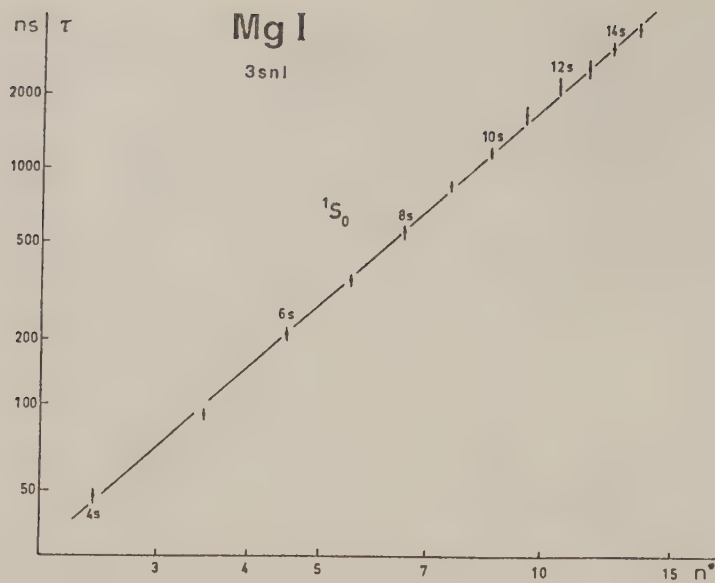


Fig. 21. Experimental lifetimes in the $1S_0$ sequence in Mg.

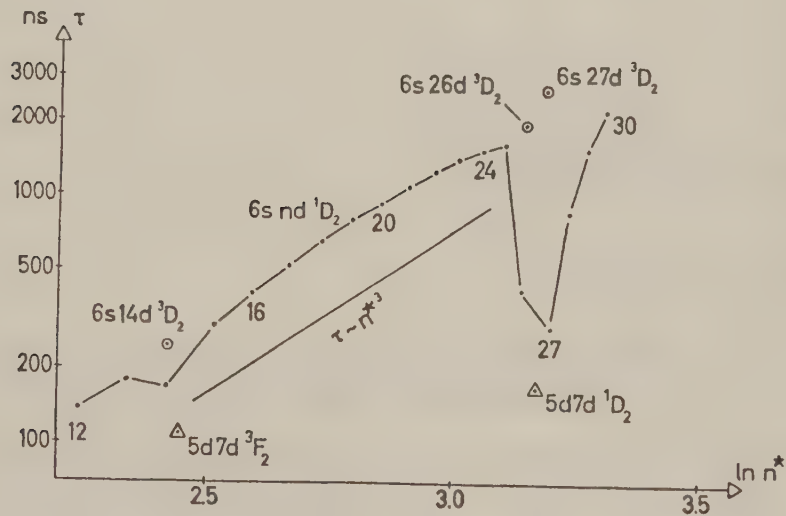


Fig. 22. Experimental lifetimes in the $1D_2$ sequence in Ba.
(From Ref. [31].)

value [30,31]. (See Fig. 22.) Lifetime values can then be used to calculate the degree of admixture.

The $1F_3$ -sequence in Sr is perturbed by the admixture of a long-lived doubly excited state. Multiconfiguration Hartree-Fock calculations have shown that about 50% of the $4d5p$ $1F_3$ state is mixed into the $5s4f$ $1F_3$ state [32]. (See Fig. 23.) Here it is difficult to verify the admixture coefficients from lifetime data since the unperturbed lifetime of the $5s4f$ $1F_3$ state is

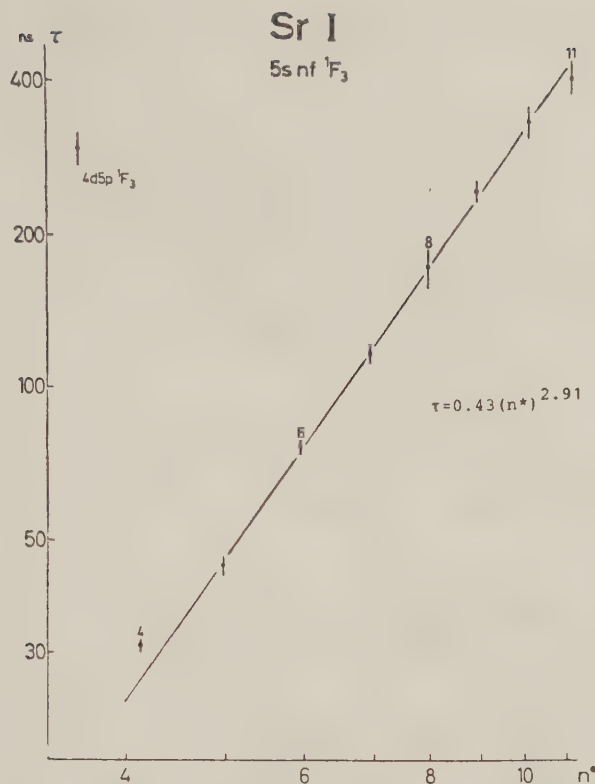


Fig. 23. Experimental lifetimes in the $1F_3$ sequence in Sr.

not known. This state is the lowest in the sequence and will not necessarily scale like the rest.

In the $1D_2$ -sequence in Mg the admixture coefficients have been determined using multiconfigurational Hartree-Fock calculations [29] and Multichannel Quantum Defect Theory (MQDT) [33]. The major perturber in the sequence is a $3p^2$ state, which is not identified as a localized energy level. The admixture is largest in the $3d\ 1D_2$ state and decreases with n . The admixture lifts the lifetime values in the lower part of the sequence. (See Fig. 24.) Our experimental results in the $1D_2$ sequence are in good agreement with theoretical calculations [29]. This leads to very well established experimental oscillator strengths for the lower members of the $1S_0$ and $1D_2$ sequences in Mg.

If a Rydberg series is perturbed by an admixture of several states or if the perturbation is smeared out the resulting lifetimes may be difficult to explain. In the $2D_{5/2,3/2}$ sequences in In an interaction with the autoionizing $5s5p^2\ 2D$

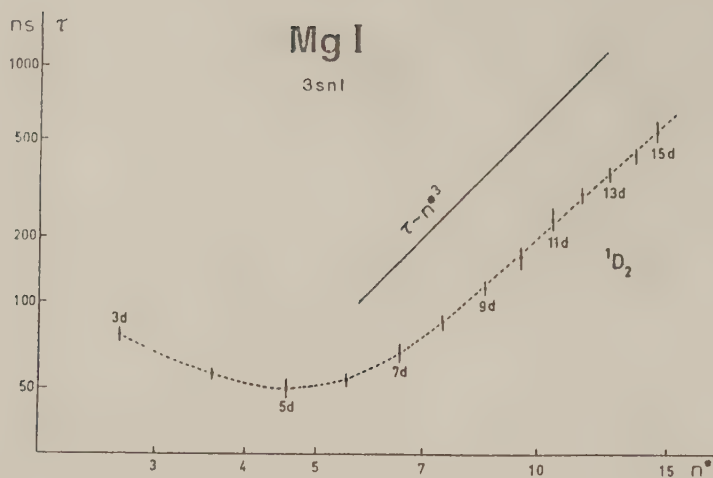


Fig. 24. Experimental lifetimes in the $1D_2$ sequence in Mg.

state has been suggested to explain the low absorption cross section for the $n=7-10$ states [34]. Here the Coulomb approximation completely fails to calculate the lifetime of the $7d \ ^2D_{3/2}$ state [28]. The corresponding sequences in Al is perturbed by the $3s3p^2 \ ^2D$ state, which lies below the first ionization limit and it has been shown that the entire 2D sequence is affected by configuration interaction [35]. In the 1P_1 sequence in Sr the behaviour can to some extent be explained by the admixture of the short-lived $4d5p \ ^1P_1$ state [36] which would decrease the lifetimes. In the 1D_2 sequence in Sr MQDT calculations have shown that the $4d6s \ ^1D_2$ perturbation is spread out over a number of levels [36]. The sequence is also perturbed by singlet-triplet mixing [36] and a more detailed analysis is needed to fully explain the behaviour.

IV.2 Hyperfine Structure

The hyperfine-structure splitting in a hydrogen-like atom is expected to scale as n^{-k} , where k is close to three. Combined with the scaling of the lifetime values this results in a spectral resolution, which should be about constant through a long sequence of a Rydberg series in a hydrogen-like atom. In the alkali atoms having no perturbing states, the scaling law of the hfs is well fulfilled. Our results from the $^2P_{3/2}$ sequence in ^{23}Na is, together with previous measurements [37], in excellent agreement with the simple scaling rule. Group III A elements have one electron outside a closed subshell with two s-electrons.

Highly excited Rydberg states are then expected to behave like corresponding states of alkali-atoms but the situation can be complicated by the presence of doubly excited states. Fig. 25 shows the magnetic dipole interaction constant in the $^2P_{3/2}$ sequence in ^{27}Al . The measured a-values scale as $(n^*)^{-2.85}$. Being a light element aluminum is well suited for calculations using Many Body Perturbation Theory. Such calculations are planned for the 2P sequence [38].

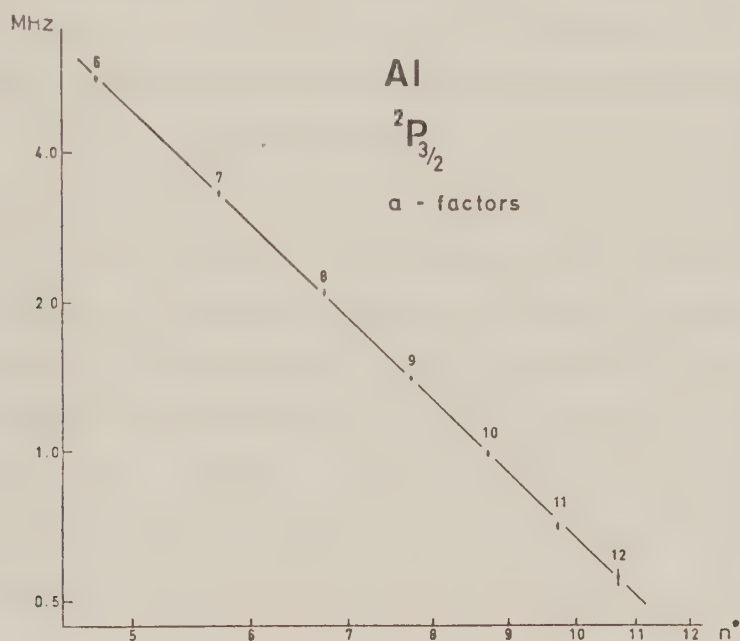


Fig. 25. Experimentally determined magnetic-dipole interaction constants in the $^2P_{3/2}$ sequence in ^{27}Al .

In the alkaline-earth atoms, having two valence electrons, the situation is more complex. In a Rydberg series with one excited electron the hyperfine structure is mainly determined by the unexcited s-electron, which has a higher probability density at the nucleus. This means that the hyperfine splittings in an unperturbed Rydberg series is nearly constant. Only a small part of the hyperfine structure, owing to the excited electron, is decreasing with n^* . If a Rydberg series is perturbed by the admixture of doubly excited states the density of the valence s-electron at the nucleus is reduced. This leads to a change in both hfs and isotope shifts. If the coupling between the excited electron and the remaining s-electron is changed both isotopes

with and without nuclear spin are affected. That is not the case for the so called hyperfine-induced singlet-triplet mixing. In highly excited alkaline-earth atoms the hyperfine structure may be of comparable size to the fine structure splitting. The result will be a recoupling of the angular momenta giving rise to a change in the hyperfine structure and a shift of the isotopes with hyperfine structure.

Measurements in the 1D_2 sequence in Ba showed a resonant behaviour of the hyperfine structure in the region where lifetime measurements previously had shown effects of the admixture of the doubly excited $5d7d\ ^3F_2$ state [31]. The isotope shifts in the sequence responded also to the admixture. The shift of the even isotope was essentially constant through the measured states whereas the shift of the odd isotopes were changing rapidly. (See Fig. 26). Together with an increase in the hyperfine structure splitting this indicates the presence of hyperfine induced singlet-triplet mixing [39]. In view of the increasingly strong hyperfine-induced singlet-triplet mixing the deduced b-factor should probably be considered as a fitting parameter for the high n-values [40]. Measurements revealing similar hyperfine induced singlet-triplet mixing has been performed on Sr and Ca [41].

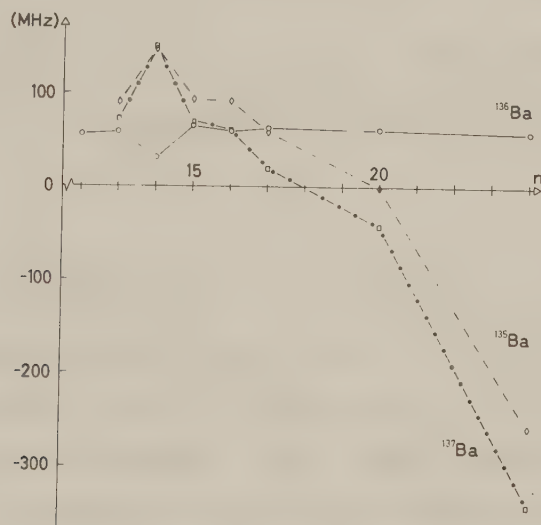


Fig. 26. Experimentally determined isotope shifts for $6s6p\ ^1P_1 - 6snd\ ^1D_2$ transitions in barium. The normal mass shifts have been subtracted.

Appendix: Units and designations

If nothing else is said in the text all the formulae (in the thesis and the papers) are written in SI-units. Some non-SI-units have been used, they are

$$1 \text{ ångström} = 1 \text{ Å} = 0.1 \text{ nm}$$

$$1 \text{ kayser} = 1 \text{ K} = 1 \text{ cm}^{-1} = 100 \text{ m}^{-1}$$

$$1 \text{ gauss} = 1 \text{ G} = 10^{-4} \text{ T}$$

Error limits in the papers are written with the uncertainty in the last digit within parenthesis.

For example

$$37.6(5) \text{ means } 37.6 \pm 0.5$$

and

$$12.3(14) \text{ means } 12.3 \pm 1.4$$

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Natural radiative lifetimes in the interacting $5snd\ ^{1,3}D_2$ sequences in Sr

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Natural radiative lifetimes have been determined in the $5snd\ ^{1,3}D_2$ sequences of strontium in the perturbed region around $n = 15$. Stepwise laser excitations and pulse modulation of a cw dye-laser beam were employed. A plot of the measured lifetime values versus the effective quantum number reveals complex configuration interaction.

I. INTRODUCTION

The alkaline-earth atoms have two electrons outside closed shells and the presence of low-lying, doubly excited states results in strong perturbations of sequences with only one excited electron. These configuration-interaction effects can be studied not only in the basic energy-level structure but are also reflected in, e.g., radiative lifetimes, hyperfine structures, and isotope shifts. For a theoretical description of these effects, the multichannel quantum-defect theory (MQDT) is usually applied.

In strontium, the energy levels of the $5snd\ ^{1,3}D_2$ sequences together with perturbing states have been measured by several authors.^{1,2,3} In Ref. 2 the data were analyzed with the use of MQDT. The g_J factors, calculated from the obtained wave functions, were found to agree very well with experimental values determined in a Zeeman-effect investigation in the perturbed region around $n = 15$.⁴ In the same region, a strong variation in the hyperfine interaction has also been found.⁵

In the present work we have measured radiative lifetimes in the $5snd\ ^1D_2$, $n = 13-22$, and $5snd\ ^3D_2$, $n = 14-17$ sequences of strontium using the PUMOLS technique (pulse modulated laser spectroscopy).⁶ Stepwise excitations of a collimated atomic beam were employed. By using a single-mode laser for the second step a selective excitation

of the isotope ^{88}Sr was performed. Lifetime values for lower-energy members of the 1D_2 sequence and some low-lying 1S_0 levels have been measured by Gornik.⁷

For radiative lifetimes, properties of the wave functions other than those for energy levels and g_J factors are important, and it is interesting to investigate how well different manifestations of perturbation can be reproduced. In recent measurements on barium regarding lifetimes⁸ and g_J factors,⁹ published MQDT wave functions¹⁰ were found to describe the radiative properties well but resulted in inadequate predictions of the g_J factors. This has resulted in a refinement in the MQDT treatment,^{9,11} leading to a much improved g_J description.

II. EXPERIMENTAL ARRANGEMENT

The experimental setup used in the present measurements was similar to that described in Ref. 8. A blue multimode cw dye laser, operating with the dye Coumarin 1, induced the 4607-Å transition from the ground state $5s^2\ ^1S_0$ to the intermediate $5s5p\ ^1P_1$ level, which has a lifetime of only 4.8 ns.¹² A single-mode ring dye laser, operating with the dye Stilbene 3, was used for the second step from the 1P_1 to the $^{1,3}D_2$ states. Both dye lasers were pumped by ion lasers with uv lines. The output beam of the single-mode dye laser was pulse-modulated by an acousto-optic modulator for delayed-coincidence measurements. The atomic beam of low collimation came from a resistively heated oven containing Sr metal. The two laser beams were made to overlap and crossed the atomic beam at about a right angle. The decay of the investigated level was detected with a cooled photomultiplier tube equipped with suitable filters. A photon, detected by the photomultiplier, was used as a "stop" signal, whereas a signal derived from the prompt pulse was used as a "start" signal for a time-to-amplitude converter. The pulses produced by this unit were fed to a multichannel analyzer (Tracor Northern TN 1710).

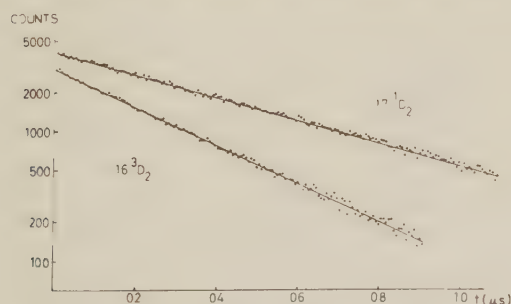


FIG. 1. Experimental decay curves for the Sr states $5s17d\ ^1D_2$ and $5s16d\ ^3D_2$. Background counts are subtracted.

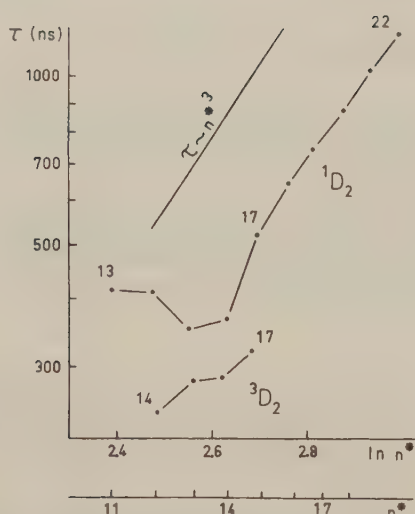


FIG. 2. Measured lifetimes in the Sr $1,3D_2$ sequences plotted vs the effective principal quantum number.

III. MEASUREMENTS AND RESULTS

In the measurements the first-step laser was tuned to the 4607-Å transition from the $5s^2 1S_0$ to the $5s5p^1P_1$ state by observing the intense blue fluorescence light. The wavelengths of the second-step laser were measured with a digital wavelength meter and were found to be in good agreement with Ref. 3. For each individual lifetime value a suitable pulse length and repetition rate was set on the acousto-optic modulator in order to get an optimum duty cycle. Since the wavelengths of the first and the

TABLE I. Experimentally determined natural radiative lifetimes for Sr states (300 K).

State	Measured τ value (ns)
$5s 13d^1D_2$	410(20)
$5s 14d^1D_2$	408(12)
$5s 15d^1D_2$	340(10)
$5s 16d^1D_2$	365(15)
$5s 17d^1D_2$	517(16)
$5s 18d^1D_2$	640(16)
$5s 19d^1D_2$	738(37)
$5s 20d^1D_2$	866(40)
$5s 21d^1D_2$	1023(51)
$5s 22d^1D_2$	1190(60)
$5s 14d^3D_2$	247(8)
$5s 15d^3D_2$	282(11)
$5s 16d^3D_2$	286(9)
$5s 17d^3D_2$	319(15)
$4d^2 3P_2$	7(2.5)
$5s 15s^1S_0$	1145(57)
$5s 16s^1S_0$	1424(72)
$5s 7f^1F_3$	126(5)
$5s 10p^1P_1$	92(5)

second step are fairly close, complete suppression of the intense blue resonance fluorescence was sometimes hard to achieve when we detected at the laser wavelength. On some occasions uv detection of the decay to the $5s5p^3P$ levels was used resulting in a more efficient suppression. Experimental decay curves for the $5s 16d^3D_2$ and $5s 17d^1D_2$ states are shown in Fig. 1. The multichannel analyzer was interfaced to a minicomputer and every decay curve was stored as a data file on a floppy disc. This provided us with the opportunity of calculating the radiative lifetime of one decay curve while another one was being stored in the multichannel analyzer. For each state, measurements were performed several times and particular attention was paid to ensure that there was no influence due to pileup, multiple scattering, collisions, Zeeman quantum beats, and flight-out-of-view effects.⁶

The experimentally determined natural radiative lifetimes for the Sr sequences $5snd^1D_2$ ($n=13-22$) and $5snd^2D_2$ ($n=14-17$) can be seen in Table I and Fig. 2. The states $5s 15s^1S_0$, $5s 16s^1S_0$, $5s 7f^1F_3$, and $5s 10p^1P_1$ could also be reached in the employed laser wavelength region and the corresponding lifetime values are included in Table I. The last two of these states were excited from the metastable $5s4d^1D_2$ level, populated in the cascade decay of the $5s5p^1P_1$ level. The given lifetime values are referred to room temperature and are not corrected for expected small effects of black-body radiation (see, e.g., Refs. 13 and 8). The lifetime of the doubly excited perturber state $4d^2 3P_2$ was also determined. In order to measure this particularly short lifetime we first recorded the shape of the laser pulse from the acousto-optic modulator by detecting reflected light inside the vacuum system. With uv detection, we recorded the time behavior of the fluorescent light from the state. With a computer program we could generate, from the pulse shape, decay curves for different lifetimes and make the best fit to our experimental curve.

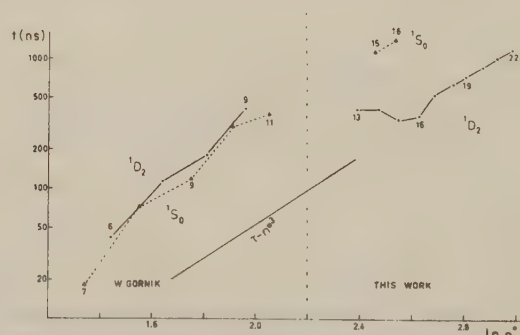


FIG. 3. Diagram including previously determined (Ref. 7) and currently measured lifetimes in the Sr $1D_2$ and $1S_0$ sequences.

IV. DISCUSSION

In Fig. 2 the lifetimes for the $^{1,3}D_2$ sequences are plotted versus the effective principal quantum number. As can be seen, there is a decrease in the lifetime values around $n=15$ for the 1D_2 sequence. In this energy range the $5snd\ ^{1,3}D_2$ levels are perturbed by the $4d6s\ ^{1,3}D_2$ levels; moreover the perturbations induce a large singlet-triplet mixing.² In the figure a line with the slope of three is also included. If the effect of perturbations is neglected, the lifetimes in a Rydberg sequence are expected to scale with a power, close to three, of the effective principal quantum number. However, to be in an overall accordance with this rule, the determined values for the $13\ ^1D_2$ and $14\ ^1D_2$ states are too long. In Fig. 3 our 1D - and 1S -state lifetime values are plotted together

with the data for lower states, investigated by Gornik.⁷ A slope-of-three line is also included. In this plot it is evident that complex perturbations are present. MQDT calculations² have shown that the $4d6s\ ^{1,3}D_2$ perturbation is spread out over a large number of Rydberg levels. It would be very worthwhile to see if the apparent shortening of the $6snd\ ^1D_2$ lifetime values for $n \geq 13$ with respect to Gornik's values can be explained by taking this mixing into account.

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Natural Radiative Lifetimes in the 1P_1 and 1F_3 Sequences of Sr I

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We have measured radiative lifetimes in the $5sn p\ ^1P_1$ ($n=5-29$) and $5sn f\ ^1F_3$ ($n=4-11$) Rydberg series in neutral strontium. The measurements were performed with single-step laser excitation starting from the ground state or from a metastable state populated by collisions. The decay photons were detected using delayed coincidence technique or a transient recorder. The presence of configuration interaction in the $5sn p\ ^1P_1$ series can be observed around $n=8$. The perturbation in the $5sn f\ ^1F_3$ series is not reflected in the behaviour of the lifetime values.

Introduction

In the alkaline-earth atoms, lifetime measurements have successfully been used to probe effects of configuration interaction. Lifetime values in a Rydberg series are expected to scale with a power close to three of the effective principal quantum number. The presence of a short-lived perturbing state can cause large deviations from this rule [1]. If a series has several perturbers or if the perturbation is spread out over a number of states the effect may be less drastic. Previous lifetime measurements show, that the $5snd\ ^1D_2$ series in Sr I is perturbed in a complex way [2, 3].

Both the 1P_1 and 1F_3 series can be reached with single-step laser excitation starting from the metastable $5s4d\ ^1D_2$ state. In an atomic beam, collisions in a discharge populate all the metastable states. CW laser light from 403 nm to 707 nm was produced using different dyes. With the PUMOLS (Pulse MODulated Laser Spectroscopy) technique we measured lifetimes in the $5sn p\ ^1P_1$ ($n=7-12$) and $5sn f\ ^1F_3$ ($n=4-11$) series [1]. The 1P_1 series can also be reached from the $5s^2\ ^1S_0$ ground state in a single step if short wavelengths can be generated. With an excimer laser system we measured the 1P_1 series up to $n=29$, corresponding to wavelengths down to 218 nm. Photons released in the decay were detected with a photomultiplier tube and a transient recorder. Some lifetime values for the lower lying states in the 1F_3 series have earlier been measured with laser

spectroscopy techniques [2, 4]. These results agree with ours. In the 1P_1 sequence four states have previously been measured [5]. Our results consistently determined with both the mentioned techniques show large deviations from the published values for $n=6-8$.

Experimental Techniques

The experimental arrangements used in this work are similar to the ones described in [1, 6]. A resistively heated oven produced an atomic beam and through collisions in a discharge the metastable $5s4d\ ^1D_2$ and $^3D_{3,2,1}$ states could be populated.

In the PUMOLS experiments an Argon- or Krypton ion laser (Spectra Physics SP 171-18 or Coherent Radiation CR 3000 K) was used to pump a dye laser (Coherent Radiation CR 599 or CR 699). To get an effective pumping of the various laser dyes either UV, violet, green or red ion laser lines were employed. With the following dyes, laser light with wavelengths from 403 nm to 707 nm was produced: Stilbene 1, Stilbene 3, Coumarin 1, Coumarin 30, Rhodamine 110 and Oxazine 1. The dye laser was not always used with a single cavity mode. If multi-mode operation was chosen, the folding mirror was piezo-electrically vibrated to get an effectively white excitation of the studied state. The dye-laser light

was modulated by an acousto-optic modulator (SORO MAR 50). It should be noticed, that the crystal in the modulator absorbs light with wavelengths shorter than 400 nm. The modulator was controlled by a pulse generator (HP-8013 B), which also provided a start pulse for the time-to-amplitude converter (TAC). The pulse length and pulse repetition rate was individually set for each studied state. The fluorescence photons passed through interference filters and were detected by a Peltier-cooled photomultiplier tube (EMI 9558 QB). The first detected photon caused a stop signal to the TAC. The decay curves were sampled in a multichannel analyzer (Tracor Northern TN1710) and the lifetime values were calculated on a mini-computer (PDP-11V03).

In the pulsed laser experiments an excimer laser (Lambda Physik EMG 102) operating with XeCl pumped a dye laser (Lambda Physik FL 2002) using Coumarin 120, Coumarin 47, Coumarin 30 or Rhodamine 6G dye. The dye laser light was frequency doubled with a KD*P- or a KPB-crystal. In this way states corresponding to wavelengths between 218 nm and 293 nm could be reached. Decay photons passed through interference filters and were detected by a fast photomultiplier tube (EMI 9816 QB). The decay signal was captured by a transient digitizer (Biomation 8100). The transients were added in a memory and transferred to the mini-computer for calculations.

Measurements and Results

Figure 1 shows the excitation scheme used in the experiment. The $5s\,nf\,^1F_3$ sequence was excited from

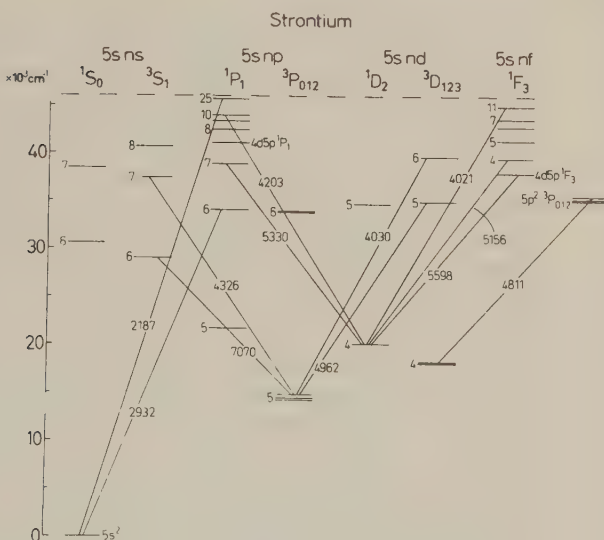


Fig. 1. Excitation scheme in the Sr I spectrum

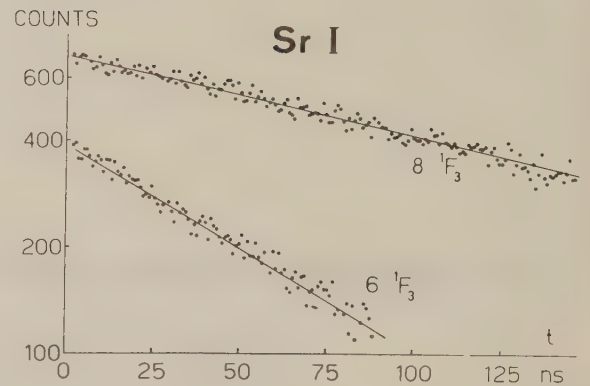


Fig. 2. Two experimental PUMOLS curves recorded with the dye laser running multi mode, drawn on a logarithmic scale. Each curve has a sampling time of about 2 min.

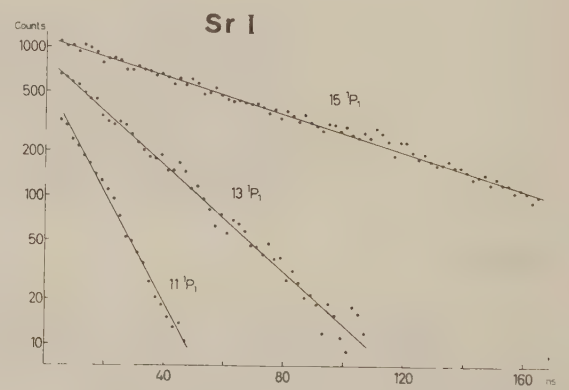


Fig. 3. Three experimental decay curves recorded with a fast transient digitizer. Each curve is the sum of about 500 transients

the metastable $5s\,4d\,^1D_2$ state. The subsequent decay was detected on the laser wavelength. Figure 2 shows two experimental PUMOLS-recordings on a logarithmic scale, obtained when the dye laser was running multi-mode. The $5s\,np\,^1P_1$ sequence was excited either from the $5s^2\,^1S_0$ ground state using the pulsed laser system or from the metastable 1D_2 state using pulse-modulated CW light. The decay of 1P_1 states to the $5s\,4d\,^1D_2$ state was the easiest to detect but on some occasions the transition to the $5s^2\,^1S_0$ state was also measured. Figure 3 shows three experimental curves on a logarithmic scale, obtained with the pulsed laser system after adding about 500 transients. Additional measurements of some triplet states was also performed. The excitation then started from the metastable $5s\,4d\,^3D$ states.

During the lifetime measurements a magnetic field was applied in the excitation volume to avoid influence of slow Zeeman quantum beats. Normal precautions were taken to avoid systematic errors due to multiple scattering or collisions. In the PUMOLS

Table 1. Experimental lifetimes in Sr I. Excitation wavelengths are included.

State	Wavelengths [Å]		Lifetimes	
	CW laser system	Excimer laser system	This work [ns]	Other work [ns]
$5s\,5p\,^1P_1$	4,607			5.12 ^a
$5s\,6p\,^1P_1$		2,932	65(5)	3.65(14) ⁵
$5s\,7p\,^1P_1$	5,330	2,569	39.2(20)	4.93(32) ⁵
$5s\,8p\,^1P_1$	4,481	2,428	27.1(13)	5.46(17) ⁵
$5s\,9p\,^1P_1$	4,313	2,354	42.7(20)	
$5s\,10p\,^1P_1$	4,203	2,307	81(6)	
$5s\,11p\,^1P_1$	4,128	2,275	125(9)	
$5s\,12p\,^1P_1$	4,076	2,253	170(20)	
$5s\,13p\,^1P_1$		2,238	250(30)	
$5s\,14p\,^1P_1$		2,218	510(35)	
$5s\,15p\,^1P_1$		2,211	690(40)	
$5s\,16p\,^1P_1$		2,206	925(55)	
$5s\,17p\,^1P_1$		2,202	1,190(90)	
$5s\,18p\,^1P_1$		2,199	1,550(120)	
$5s\,19p\,^1P_1$		2,196	1,890(180)	
$5s\,20p\,^1P_1$		2,194	2,370(210)	
$5s\,21p\,^1P_1$		2,192	2,730(260)	
$5s\,23p\,^1P_1$		2,189	4,270(430)	
$5s\,25p\,^1P_1$		2,187	5,650(500)	
$5s\,29p\,^1P_1$		2,184	7,500(900)	
$4d\,5p\,^1F_3$	4,756	2,429	23.4(23)	
$5s\,4f\,^1F_3$	5,156		31.3(9)	29.7(9) ⁴ , 33.5(12) ² , 34.2(4) ¹³ , 34.3(28) ¹²
$5s\,5f\,^1F_3$	4,678		45.0(21)	33.6(13) ² , 53(2) ⁴
$5s\,6f\,^1F_3$	4,406		78(3)	81(8) ⁴ , 98.5(12) ²
$5s\,7f\,^1F_3$	4,253		120(5)	133(4) ⁴
$5s\,8f\,^1F_3$	4,159		179(16)	228(8) ⁴
$5s\,9f\,^1F_3$	4,096		255(13)	
$5s\,10f\,^1F_3$	4,052		350(27)	
$5s\,11f\,^1F_3$	4,021		430(35)	
$4d\,5p\,^1F_3$	5,598		296(22)	
$5s\,6s\,^3S_1$	7,070		15.0(8)	10.9(11) ¹²
$5s\,7s\,^3S_1$	4,326		34.2(17)	34.8(13) ²
	4,438			
$5p^2\,^3P_1$	4,742		8.3(4)	8.8(12) ¹² , 10.2(19) ²
	4,784			
	4,876			
$5p^2\,^3P_2$	4,811		8.3(4)	7.8(12) ¹¹ , 7.89(5) ¹³
$5s\,5d\,^3D_2$	4,872		16.0(6)	16.34(13) ¹³
$5s\,5d\,^3D_3$	4,962		15.9(6)	16.29(24) ¹³ , 17.1(8) ⁴
$5s\,6d\,^3D_2$	4,032		32.0(15)	
$5s\,6d\,^3D_3$	4,030		32.2(16)	34.0(31) ⁴

^a Weighted mean from [5] and Refs. therein

experiments the laser light sometimes had to be attenuated to avoid pile-up in the strong transitions. The time scale corresponding to different TAC ranges were determined using a time delay generator (Berkeley Nucleonics Corp. 7030). The transient digitizer was calibrated with a radio-frequency signal and the channels are separated by 10 ns. The results from our measurements are collected in Table I. Each state was measured on several oc-

casions. Lifetimes for states measured with both the laser systems are weighted mean values of all the obtained experimental curves. The lifetime values pertain to room temperature and the influence of black body radiation [7] has not been compensated for. The stated errors include both statistical scattering and estimated possible systematic errors. Included in Table I are also excitation wavelengths [8] and measurements made by other authors. Our lifetime values are in good agreement with literature values, except for the 6–8 1P_1 states, previously obtained using the Hanle method [5]. The origin of the large deviations of these lifetime values is unknown⁸. We also measured the lifetime of the $5s\,5p\,^1P_1$ state. Our result was consistent with earlier established values, but with a larger error due to influence of the excitation pulse.

Discussion

If perturbations are neglected, the radiative lifetimes in a Rydberg series⁹ are expected to scale with a power, close to three, of the effective principal quantum number. The $5snf\,^1F_3$ series, shown in Fig. 4, interacts strongly at $n=4$ with the doubly excited

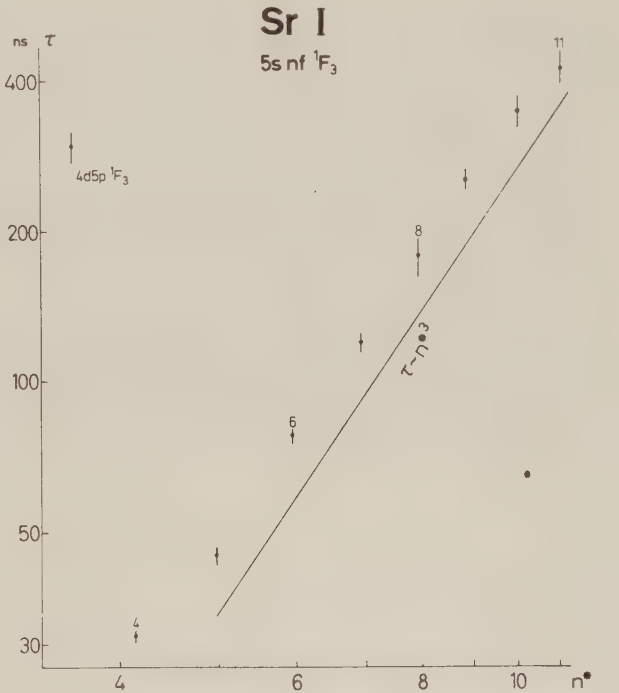


Fig. 4. Experimental lifetimes for the $5snf\,^1F_3$ ($n=4-11$) sequence in strontium plotted versus the effective principal quantum number. The diagram is drawn on a log-log scale. The $4d\,5p\,^1F_3$ perturbing state is included. The error bars are indicated by vertical lines. The drawn line indicates the slope in case of an $(n^*)^3$ dependence

⁸Footnote

Our lifetime values are consistent with results obtained from emission spectroscopy measurements [9].

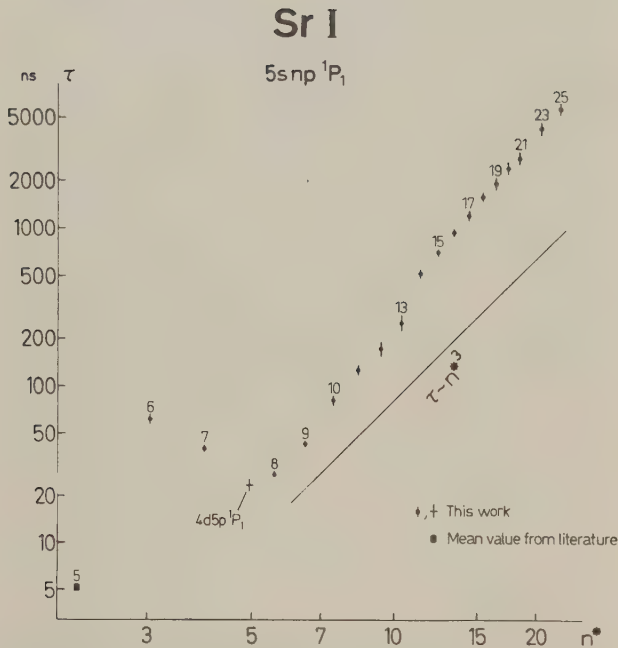


Fig. 5. Experimental lifetimes for the $5snp\ ^1P_1$ ($n=5-25$) sequence in strontium plotted versus the effective principal quantum number. The lifetime value for the $5\ ^1P_1$ state is taken from [5]. The $4d5p\ ^1P_1$ perturbing state is included

state $4d5p\ ^1F_3$. Multiconfigurational Hartree-Fock calculations predict a configuration mixing between the $4d5p\ ^1F_3$ and $5s4f\ ^1F_3$ states close to 50% [10]. A large difference in radiative lifetime between the two states is predicted due to extensive cancellation in the dipole transition from the $4d5p\ ^1F_3$ state to the lowest 1D_2 state. This is in good agreement with our observations. A fit to our experimental data yields $\tau = 0.43(n^*)^{2.91}$ for the rest of the 1F_3 series ($n=5-11$).

The $5snp\ ^1P_1$ series, shown in Fig. 5, is perturbed by the doubly excited $4d5p\ ^1P_1$ state. This state has a shorter lifetime than all the $5snp\ ^1P_1$ states with $n > 5$ and is energy-wise located between the $5s7p$

1P_1 and $5s8p\ ^1P_1$ levels. Calculations based on multichannel quantum defect theory have shown, that the $4d5p\ ^1P_1$ perturber is spread out over a large number of the low-lying 1P_1 states [11]. For lower n -values this interaction may explain most of the deviation from the simple scaling law.

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Natural Radiative Lifetimes and Stark-Shift Parameters in the $4p^2$ Configuration in Ca I

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Abstract

The radiative lifetimes in the doubly excited $4p^2$ configuration in Ca were determined using the delayed coincidence technique. The measured states were populated either with two-step laser excitation from the $4s^2\ ^1S_0$ ground state or with single-step excitation starting from a metastable state. The metastable states were populated by collisions in a discharge. The following lifetime values were obtained: $\tau(4p^2\ ^1D_2) = 16.3(20)$ ns, $\tau(4p^2\ ^1S_0) = 14.4(7)$ ns and $\tau(4p^2\ ^3P_{0,1,2}) = 6.9(4)$ ns. Stark shift parameters in the same configuration were measured using an atomic beam passing between electric field plates. The following results were obtained: $\alpha_0(4p^2\ ^1S_0) = -32.2(16)$ MHz (cm/kV) 2 , $\alpha_0(4p^2\ ^1D_2) = 1.8(2)$ MHz (cm/kV) 2 and $\alpha_2(4p^2\ ^1D_2) = 1.15(10)$ MHz (cm/kV) 2 . Additional measurements yielded $\tau(4s5f\ ^1F_3) = 60(3)$ ns, $\tau(4s4d\ ^3D_{1,2,3}) = 13.0(5)$ ns and $\alpha_0(4s6s\ ^1S_0) = 6.98(35)$ MHz (cm/kV) 2 .

1. Introduction

Configuration interaction with doubly excited states in sequences of Rydberg levels is a prominent feature in the alkaline earth elements (See, e.g., [1].) Such configuration interaction and associated singlet–triplet mixing can be probed in systematic measurements of a number of atomic parameters such as natural radiative lifetimes, Stark interaction constants, Landé factors and hyperfine structure constants. Multichannel quantum defect theory is well suited for describing perturbations of this kind (See, e.g., [2].) Because of the large difference in lifetimes for a typical Rydberg state (μ s) and a doubly excited state (ns), even small admixtures into Rydberg states can be clearly noticed. In a similar way, Rydberg states are very sensitive to external electric fields and have very high polarizabilities. Thus, in a Stark-effect investigation on a doubly excited state, admixtures of Rydberg character will be observed as a strongly increased polarizability. We have previously investigated the lifetime behaviour in sequences of Ba [3, 4] and Sr [5, 6] states. In the present paper we present lifetime results for the short-lived doubly excited $4p^2$ configuration in Ca. We have also measured Stark interaction parameters in this configuration. There is a close connection between natural lifetimes and Stark parameters. The theoretical calculations of radiative properties and Stark polarizabilities all involve the evaluation of matrix elements of the electric dipole operator [7].

Lifetime measurements have been much facilitated through the selective state excitation that is possible with pulsed tunable lasers, enabling direct observation of the exponential decay without cascade influence. The fluorescence light can for instance be captured by a transient recorder and after adding several decays the lifetime can be calculated from the exponential curve. If the repetition rate of the laser pulses is high, single photons can be detected in delayed-coincidence experiments, that normally result in precision values. However, both methods

run into problems if the studied lifetime is short compared with the risetime of the laser pulse. The fluorescence curve is then, because of the influence of the laser pulse, not a pure exponential. In the present laser-excitation work the short-lived doubly excited $4p^2$ Ca states pose problems of this kind. In order to improve our lifetime measurements for short-lived states we have recorded the laser pulse shape and fitted computer generated fluorescence curves to our experimental data. We also investigated the possibility of modulating the CW laser light sinusodially and calculating the lifetime from the observed phase-shift (See, e.g., [8]).

In all atoms except hydrogen, the Stark interaction in a “weak” electric field results in energy level shifts, that are proportional to the square of the field strength. Low-lying states are only little affected whereas the loosely bound highly excited states easily are driven into a linear hydrogen-like region, ultimately leading to field ionization. Extensive Stark effect investigations have been performed for the alkali atoms both in the “low”-field regime (See, e.g., [9–11]) and in the “high”-field regime (See, e.g., [12, 13]). Some investigations have also been performed for the alkaline earth elements. Thus, the perturbation in the $6snd\ ^1D_2$ sequence of Ba close to $n = 26$ was studied in this way by Gallagher et al. [14]. Similar measurements were performed around the $4d6s$ perturbation in Sr $1,3D_2$ states by Gessert et al. [15]. van Leeuwen and Hogervorst have investigated several low-lying Ba states [16] and the $4sns\ 3S$ and $4snd\ ^3D$ Rydberg sequences of Ca [17]. In [16] they also included the $3d^2$ doubly excited Ca configuration, for which the Stark interaction provided an extremely sensitive configuration interaction probe. The presently investigated $4p^2$ doubly excited configuration is of lower energy, and increased difficulties in the theoretical interpretation can be anticipated [16].

2. Experimental techniques

Figures 1(a, b) show the three different experimental setups used in the present work. The experiments were all performed on an atomic beam of Ca produced in a vacuum chamber. The fluorescence light passed through interference filters and was detected by a Peltier-cooled photomultiplier tube (EMI 9558 QB).

The studied states were excited in two steps. The excitation scheme is shown in Fig. 2. We used either two-step laser excitation or single-step excitation starting from a metastable state. The metastable states were populated by collisions in a discharge [18, 19].

Figure 3 shows the oven used for metastable atom production. A typical discharge current was about 100 mA using 20 V potential difference. The first step laser excitation used

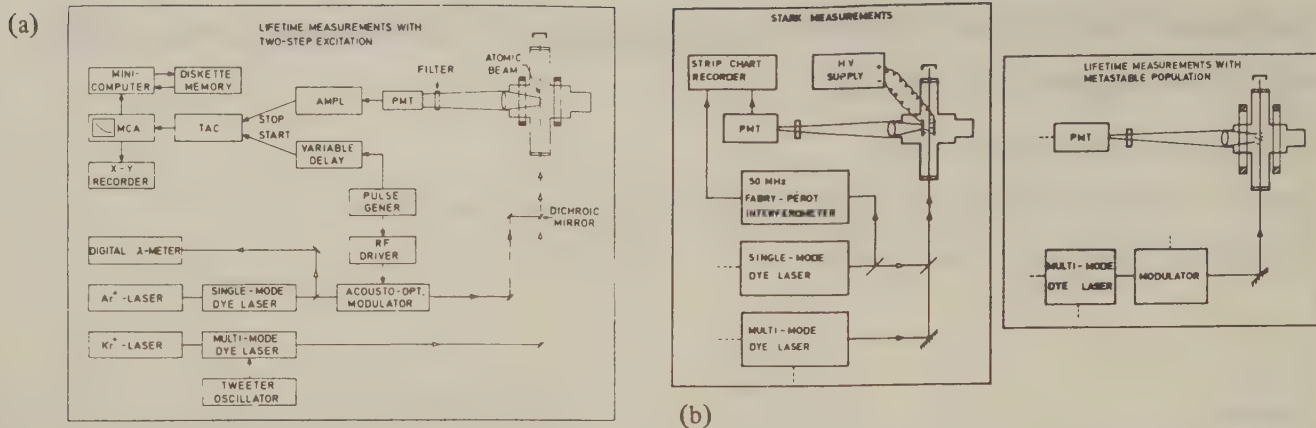


Fig. 1. (a, b) Experimental set-ups used in the lifetime and Stark shift measurements.

a UV-pumped multi-mode dye laser operating on Stilbene 3 dye (Coherent Radiation CR 3000K and CR-599). The piezo-mounted folding mirror in the dye laser was vibrated to achieve an effectively white excitation of the intermediate $4s4p^1P_1$ state. The second step was excited with a single-mode dye laser system operating on Rhodamine 6G dye (Spectra Physics 171-17 and CR-599-21). In this way the $4p^2^1S_0$ and 1D_2 states were investigated. Using metastable atoms the laser excitation started from the $4s3d^1D_2$ or the $4s4p^3P_{2,1,0}$ states. The multi-mode dye laser system was then used. The $4p^2^3P_{2,1,0}$ states and the additional states $4s4d^3D_{3,2,1}$ and $4s5f^1F_3$ were excited from metastable states.

During the lifetime measurements the second step-laser light was modulated by an acousto-optical modulator (SOROMAR-50). Using a controlling pulse generator (HP 8013), square-wave light pulses with suitable length and pulse rate were formed. From the exciting pulse a start signal for the time-to-amplitude converter (TAC) was also derived. One state was studied with the laser light modulated in a sinusoidal manner. The first photon detected by the PMT caused a stop signal for the TAC. Pulses, with different heights, coming from the TAC

were collected in a multichannel-analyzer (Tracor Northern TN 1710). The decay curves were transferred to a minicomputer (PDP-11V03) for calculation of the lifetimes. The basics of the lifetime measurement technique (PUMOLS, Pulse-MODulated Laser Spectroscopy) have been described in [20].

In the Stark-shift measurements the atomic beam was collimated with apertures and passed between condensor plates generating a homogeneous static electric field. Electric field strengths up to 12 kV cm^{-1} could be used. Part of the second step laser light was sent to a 50 MHz multipass Fabry-Pérot interferometer. The Fabry-Pérot fringes were recorded together with the signal from the studied state. If possible we performed recordings with and without electric field in the same laser scan. During the measurements the electric field was monitored with a high-voltage meter (Hickok 3301).

3. Measurement and results

During the lifetime measurements a magnetic field was applied in the excitation volume to avoid influence of slow Zeeman quantum beats. The square wave pulse modulation was individ-

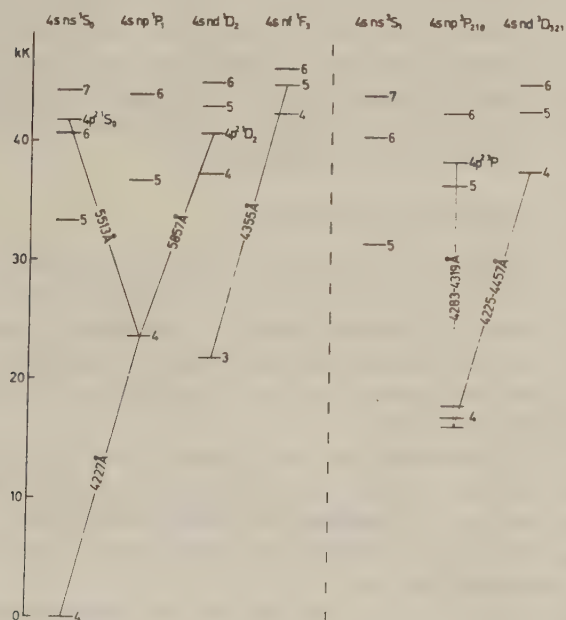


Fig. 2. Excitation scheme in the Ca I spectrum.

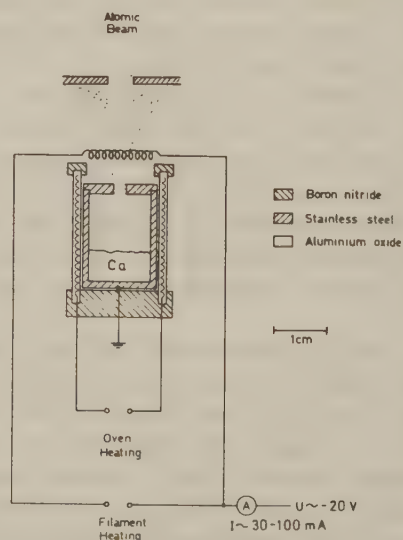


Fig. 3. Principal drawing of the metastable oven. The height of the oven is about 3.5 cm.

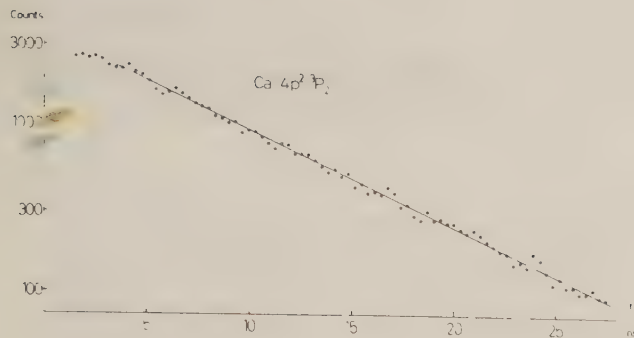


Fig. 4. An experimental decay curve from the short-lived $4p^2 {}^3P_2$ state in Ca. The straight line would correspond to a lifetime about 5% longer than the one derived taking the laser-pulse shape into account.

ually set for each studied state. The lifetime values were normally derived directly from the exponential decay curves. However, for very short lifetimes the modulation of the laser pulse can not be regarded as a perfect square wave. The laser pulse shape was then recorded from light scattered on a rod, that was moved into the excitation region. From the laser pulse computer-generated fluorescence curves corresponding to different lifetimes were fitted to our experimental decay data. Figure 4 shows an experimental recording of the shortlived $4p^2 {}^3P_2$ state, on a logarithmic scale. In this case, the slope of the straight line only sets an upper limit for the lifetime value, due to influence of the excitation light. Calculations using the laser pulse shape will produce the accurate value.

In Fig. 5 the difference in decay curve behaviour when passing from a case of a long to a short lifetime τ is illustrated when working with the PUMOLS technique.

We see that the technique gradually can be transformed into the phase-shift method [8, 21]. We then deliberately smooth the leading and trailing ends of the pulses by performing a sinusoidal modulation at the frequency Ω . If the exciting wave I_{exc} is expressed as [21]

$$I_{\text{exc}} = I_0(1 + a \sin \Omega t) \cos \omega t \quad (1)$$

the fluorescence light I_{fl} will feature a modulation with a phase shift φ and a reduced contrast $a/\sqrt{1 + \Omega^2 \tau^2}$ according to

$$I_{\text{fl}} = bI_0 \left[1 + \frac{a}{\sqrt{1 + \Omega^2 \tau^2}} \sin(\Omega t + \varphi) \right] \cos \omega t \quad (2)$$

with $\tan \varphi = \Omega \tau$.

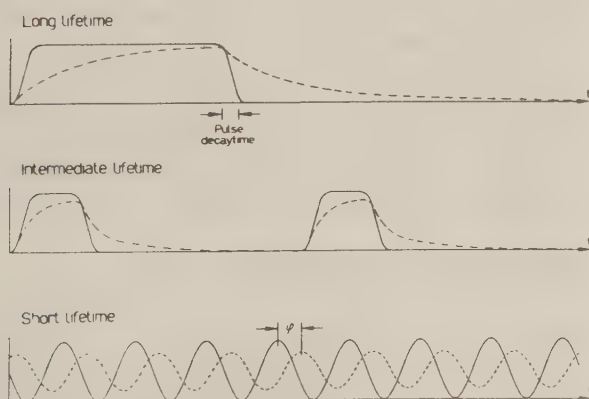


Fig. 5. Illustration of measurements of long and short lifetimes, with gradual transition from pure exponential decay evaluation to phase-shift determination.

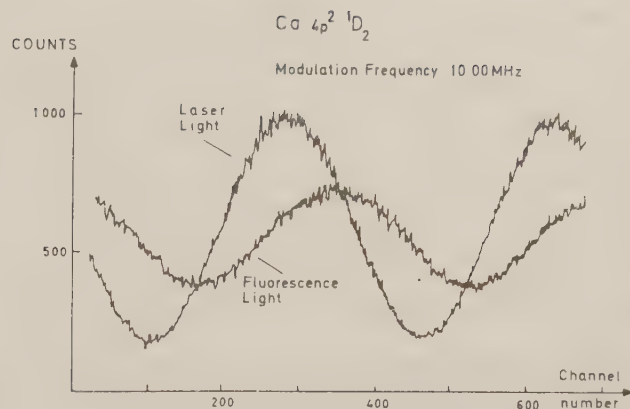


Fig. 6. A recording of the laser light and the corresponding fluorescence from the Ca $4p^2 {}^1D_2$ state, when the laser was modulated sinusoidally. The phase shift in this recording was calculated to be 46° .

The $4p^2 {}^1D_2$ state was measured with the phase-shift version of PUMOLS. Figure 6 shows a recording of both the laser light and the corresponding fluorescence.

Each state was measured several times and on several occasions. Normal precautions were taken to avoid systematic errors due to multiple scattering or collisions [20]. The results from the lifetime measurements are collected in Table I. The fine structure levels in the $4p^2 {}^3P$ and $4s4d {}^3D$ states were investigated separately but no J -dependence was found. Our lifetime values are in good agreement with previous measurements [18, 22, 23].

The Stark shift ΔT_m of a m -sublevel with total angular momentum J is given by

$$\Delta T_m = -\frac{1}{2} \left[\alpha_0 + \alpha_2 \frac{3m^2 - J(J+1)}{J(2J-1)} \right] E^2 \quad (3)$$

where E is the electric field strength and α_0 and α_2 are the scalar and tensor polarizability constants, respectively. 1S_0 states having $J=0$ will only be shifted when an electric field is applied whereas states with $J \geq 1$ will be both shifted and split up into more than one component. The shift in a transition between two states will be affected by the polarizabilities of both involved states. In our case influence of the intermediate $4s4p {}^1P_1$ state can be neglected [24, 25] compared to the shifts in the measured states.

The linewidth of the signals is determined by residual Doppler broadening and by the short lifetime 4.7(5) ns [23] of the intermediate state. Figures 7 and 8 show recordings of two transitions to 1S_0 states. The peak positions with and without electric field were recorded in the same laser scan. In this way

Table I. Calcium lifetimes

State	Lifetime (ns)	
	This work	Other work
$4p^2 {}^1S_0$	14.4(7)	12.6(8)[23] ^b
$4p^2 {}^1D_2$	16.3(20) ^a	14.9(9)[23], 16.1(8)[22]
$4p^2 {}^3P_{2,1,0}$	6.9(4)	
$4s4d {}^3D_{3,2,1}$	13.0(5)	12.1(7)[18] ^c
$4s5f {}^1F_3$	60(3)	

^a The result is obtained with the phase-shift version of PUMOLS.

^b Previously identified as $4s6s {}^1S_0$, interchanged in [26].

^c 3D_1 .

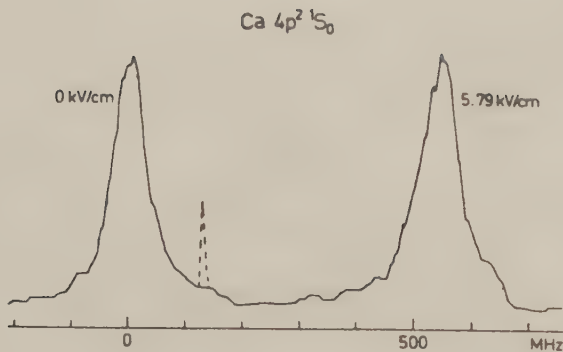


Fig. 7. Stark-shift recording of the $4s4p\ ^1P_1-4p^2\ ^1S_0$ transition. The two peaks, without and with electric field, were obtained in one laser sweep. The sharp spike is due to the Stark shifted level sweeping through resonance when the electric field is switched on.

systematic errors due to possible mode-hops and drifting of the Fabry-Pérot fringes are minimized. Figure 9 shows recordings of the $4s4p\ ^1P_1-4p^2\ ^1D_2$ transition. The tensor structure is not fully resolved. Due to the small shifts the recordings with and without electric fields were done separately. The three studied transitions were measured several times and with different electric fields. The square dependence of the electric field was verified ensuring the validity of the "weak"-field approximation, eq. (3). The calculated Stark-shift parameters are collected in Table II. The statistical scattering in our measurements is small and the stated errors are of possible systematic nature, e.g., uncertainties in the separation of the electric field plates.

4. Discussion

Radiative lifetimes in short lived excited states ($\tau < 10$ ns) can be measured accurately with the PUMOLS technique as first illustrated in the present paper. It is then necessary to record the exciting laser pulse and to computer generate fluorescence curves and fit them to the non-exponential decay curves. Short-lived states can also be measured using the phase-shift method. An acousto-optic modulator is very non-linear in the driving field and reasonable sinusoidal modulation could only be obtained at certain frequencies which limited the applicability in our experiments. However, in principle the phase-shift of the first Fourier component could be used for an arbitrary modulation [21].

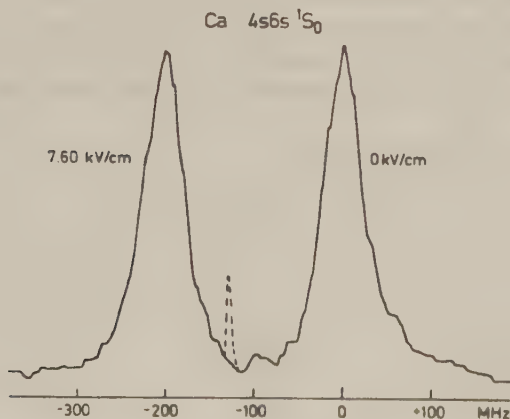


Fig. 8. Stark-shift recording of the $4s4p\ ^1P_1-4s6s\ ^1S_0$ transition. The peaks were recorded in one laser sweep. The sharp spike indicates the switching off of the electric field.

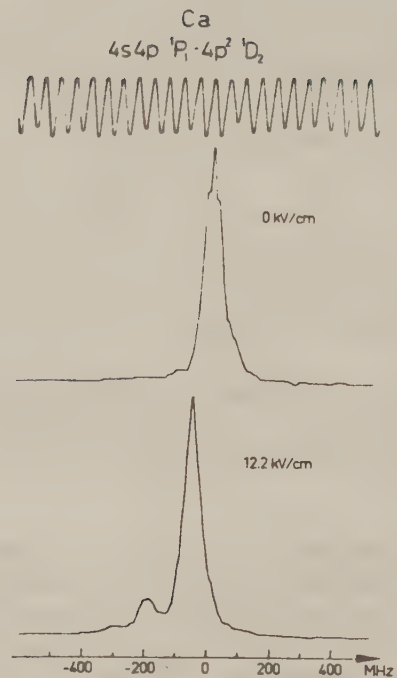


Fig. 9. Stark shift recording of the $4s4p\ ^1P_1-4p^2\ ^1D_2$ transition. The fringes from the 50 MHz Fabry-Pérot interferometer are included.

Since the temporal shape of the fluorescence light can be calculated with τ as a parameter for any excitation function it is clearly not necessary to identify an experimental phase-angle φ for deriving the lifetime τ . Thus, it is only a matter of terminology if we use the expression "phase-shift method" or not.

The doubly excited $4p^2$ states were found to be shortlived as expected. In particular, the previously not measured $^3P_{2,1,0}$ states have very short lifetimes. The agreement with the measurement of Havey et al. [23] in the case of the $4p^2\ ^1S_0$ and 1D_2 states is very satisfactory. These authors also measured several other low-lying Ca states, in particular the $4sns\ ^1S_0$ levels for $n = 6, 7$ and 8 ($\tau = 88.9(30), 62.4(43)$ and $118.3(63)$ ns, respectively). The influence of the perturbing $4p^2\ ^1S_0$ state on the lifetimes is very clear in this sequence for which the unperturbed lifetimes should increase with $(n^*)^3$ (See, e.g., [4].) The lifetime values for the $4p^2\ ^1S_0$ and $4s6s\ ^1S_0$ states strongly support the level designations proposed by Risberg [26], which have been adopted in the present work in contrast to [23].

In [16] a theoretical interpretation of measured polarizability constants for the $6p^2\ ^3P$ states is attempted employing dipole matrix elements evaluated from Coulomb-approximation and configuration-interaction wavefunctions. Agreement between theory and experiment was found to be poor due to shortcomings of both theoretical approaches for states of intermediate energy. The $4p^2$ configuration is the corresponding one in Ca and it could be anticipated that more accurate wavefunctions for the involved states than presently available would be needed for a successful interpretation.

Table II. Stark-shift parameters

State	α_0 [MHz/(kV/cm) ²]	α_2 [MHz/(kV/cm) ²]
$4p^2\ ^1S_0$	-32.6(16)	-
$4p^2\ ^1D_2$	1.8(2)	1.15(10)
$4s6s\ ^1S_0$	6.98(35)	-

Acknowledgement

This work was supported by the Swedish Natural Science Research Council.

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Natural Radiative Lifetimes in the $3sns\ ^1S_0$ and
 $3snd\ ^1D_2$ Sequences of Magnesium.

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Abstract

Natural radiative lifetimes have been determined in the $3sns\ ^1S_0$ ($n=4-15$) and the $3snd\ ^1D_2$ ($n=3-15$) sequences of Mg I employing a pulsed laser system. The radiative lifetimes in the 1S_0 sequence and of the higher members in the 1D_2 sequence are unperturbed and scale as $n^{2.6}$ (n^* is the effective principal quantum number), while the lifetimes of the lower members in the 1D_2 sequence are perturbed by the doubly excited $3p^2$ configuration. The measured lifetimes are compared with available theoretical calculations and a very good agreement is obtained.

March 1984

Introduction

The main perturbers of the Rydberg series in the heavier alkaline earth atoms Ca, Sr and Ba are states belonging to doubly excited configurations with one of the excited electrons in a low-lying, normally empty d-shell within the electronic core. The main features of the perturbations in these Rydberg states could to a large extent be parametrized and treated in a unified way using multi-channel quantum defect theory (MQDT)¹. Lately a considerable amount of new information about the interactions in these Rydberg series has been obtained in studies of Landé factors, lifetimes and hyperfine-induced singlet-triplet and n-mixing, see e.g. Refs. 2 - 5. This has resulted in a refinement of the MQDT theory.

The lighter alkaline earth atoms Be and Mg have no empty d-shells within the electronic core. The main perturbers of the singly excited, even parity Rydberg series belong to the $2p^2$ and $3p^2$ configurations, respectively. In the present investigation, natural radiative lifetimes of the $3sns\ ^1S_0$ ($n=4-15$) and the $3snd\ ^1D_2$ ($n=3-15$) sequences have been determined. The measurements were performed on a beam of Mg atoms emanating from a resistively heated oven. A pulsed laser system was used for excitation. Time-resolved detection was employed in registering the fluorescence. Two-step excitation, two-photon absorption or one-step excitation of metastable atoms, produced in a discharge in the atomic beam, was used for populating the investigated states.

Beigang and coworkers have recently measured energy level positions and hyperfine structures in Mg⁶. References to earlier work on Mg can be found in a monograph on Mg I through Mg XII⁷. Schaefer has measured the lifetimes for several states in the $nd\ ^1D_2$ sequence using delayed coincidence technique in a pulsed hollow-cathode discharge⁸. Several other authors have also performed lifetime measurements in Mg I (see Ref. 9,10 and references therein)

Experimental techniques

The set-up used in the present investigation is shown in Fig 1. In the measurements we used one or two dye lasers, depending on the excitation scheme. The dye lasers were pumped by a Lambda Physik EMG 102 E excimer laser operating with XeCl at 308 nm. The excimer laser produced 100 mJ per pulse at 10 Hz repetition rate. When performing two-step excitation, about 10% of the excimer laser light was used to pump a homebuilt Littman cavity dye laser, operating on Coumarin 153 dye. Yellow light, with a pulse energy of approximately 1 mJ, was frequency doubled in a KD^*P -crystal to yield the 285 nm UV light necessary for the first step. The second step tunable radiation was obtained from a Lambda Physik model FL 2002 dye laser, operating on TBS, Stilbene 3, Coumarin 120 or Coumarin 47 dye. The two beams were spatially and temporally overlapped in a beam of magnesium atoms formed in a vacuum system.

In the two-photon absorption experiments the light from the FL 2002 dye laser alone was focused in the atomic beam in order to populate low-lying 1S_0 - and 1D_2 -levels. Excitation from the metastable $3s3p\ ^3P$ states, populated by a discharge in the atomic beam, was also performed with this dye laser. A pair of Helmholtz coils generated a static magnetic field of about 2 mT in the excitation volume to assure that the frequency of the produced Zeeman quantum beats was beyond the high frequency cut off limit of the detection system. The fluorescence light passed through appropriate colored glass and interference filters and was detected by an EMI 9816 QB photomultiplier tube. The optical transients were captured by a Biomation 8100 transient digitizer and the signals were added in a storage memory in order to increase the signal-to-noise ratio. The added signals were transferred to a floppy disk and a nonlinear least-squares fit to an exponential was performed in a PDP-11 V03 computer.

Measurements and Results

Fig. 2 shows a schematic energy level diagram for Mg I. Excitation wavelengths were calculated from Ref.7. Wavelengths for the $3sns\ ^1S_0$ states above $n=10$ were calculated assuming a constant quantum defect in the 1S_0 sequence. Generally, two-

step excitation via the shortlived $3s3p\ ^1P_1$ state ($\sim 2\text{ ns}^9$) was employed. For the four lowest states, two-photon absorption from the ground state was used. Populating the metastable $3s3p\ ^3P_1$ level in an electric discharge and exciting on the $\lambda=4621\text{ \AA}$ intercombination line yielded consistent results for the lifetime of the $3s4s\ ^1S_0$ state. In the measurements on the higher 1S_0 states, detection could be made in the cascade fluorescence from the $3s4p\ ^1P_1$ - $3s^2\ ^1S_0$ transition as well as on the laser wavelength. The influence on the recorded signal from the lifetime of the $4p\ ^1P_1$ state ($\sim 10\text{ ns}^9$) can be disregarded. For the 1D_2 states, detection via the $4p\ ^1P_1$ state turned out to be very disadvantageous. This is in agreement with calculations by Froese Fischer¹¹, where the transition probabilities from $nd\ ^1D_2$ to $4p\ ^1P_1$ for $n=3-8$ is one or two orders of magnitude lower than from $nd\ ^1D_2$ to $3p\ ^1P_1$. However, for the $ns\ ^1S_0$ states transition probabilities to $3p\ ^1P_1$ and $4p\ ^1P_1$ are of comparable size¹². For the $4s\ ^1S_0$ and $3d\ ^1D_2$ states detection was made at the $3s3p\ ^1P_1$ - $3s^2\ ^1S_0$ resonance transition. By lowering the density in the atomic beam it was assured that multiple scattering did not affect the lifetimes. In the same way it was controlled that the lifetimes of the long-lived 1S_0 states were not affected by collisions. Other possible errors and disturbances such as flight-out-of-view effects and rf noise from the excimer laser were taken care of as in previous measurements^{13,14}. A more thorough discussion of possible systematic errors in lifetime measurements can be found in Ref. 15.

Every state was measured at several different occasions. Generally between two and four recordings for each state were taken at every occasion. Between 500 and 2000 transients were normally appropriate for obtaining a good signal-to-noise ratio. In Fig. 3 three typical experimental curves are shown. For the $5s\ ^1S_0$ state two-photon absorption was used. Our lifetime values together with excitation wavelengths and method of excitation are given in Table I. The stated error limits include both statistical scattering and an estimation of possible systematic influences. The lifetimes of the 1S_0 states above $n=10$ can possibly be affected by blackbody radiation induced transitions¹⁶, but due to good fitting with the rest of the 1S_0 sequence this effect is not expected to be more than a few per cent.

At high atomic beam densities several transitions could be observed around the $3p\ ^1P_1$ - $10d\ ^1D_2$ and the $3p\ ^1P_1$ - $11s\ ^1S_0$ transitions. These signals were comparable in strength to the $3p\ ^1P_1$ - $11s\ ^1S_0$ transition but had decay times similar to that of the $10d\ ^1D_2$ level. They also required the presence of both laser beams. A possible cause of these signals is laser enhanced formation of Mg_2 dimers in an excited state and subsequent excitation to still higher states. Two Mg atoms in their ground state can collide and form a Mg_2 molecule in its $^1\Sigma_g^+$ ground state¹⁷. For this state the potential curve is very shallow and the energy of the molecule is only some 400 cm^{-1} less than the energy of two free $3s^2\ ^1S_0$ Mg atoms. However, in a collision between a Mg atom excited to $3s3p\ ^1P_1$ by the first step laser and a ground state Mg atom the formation of the $A\ ^1\Sigma_u$ state of the molecule is an energetically very favourable process¹⁷. The formation of the $A\ ^1\Sigma_u$ state after excitation with the first step laser is also supported when observing the fluorescence from the $3s3p\ ^1P_1$ state when the second step laser is blocked. Although the lifetime of the 1P_1 state is $\sim 2\text{ ns}$, fluorescence could still be observed after $\sim 100\text{ ns}$. This then would originate from the $A\ ^1\Sigma_u$ molecules returning to the $^1\Sigma_g^+$ ground state. The effect described above was not observed when two-photon absorption was used to populate the $nd\ ^1D_2$ and $ns\ ^1S_0$ states⁶ and thus our lifetime measurements of the $4s\ ^1S_0$ and $3d\ ^1D_2$ state employing resonance-line detection are unaffected.

Discussion

In Fig. 4 a diagram of the experimental lifetimes τ versus n^* is shown on a logarithmic scale for the 1S_0 and 1D_2 sequences. A line corresponding to a $(n^*)^3$ dependence of the lifetimes, expected for a hydrogen-like case, is included. For the 1S_0 sequence, a least-squares fit yields $\tau = 4.22 (n^*)^{2.62}$ and in the 1D_2 sequence we obtain $\tau = 0.45 (n^*)^{2.65}$ for $n=8-15$. The lifetimes in the 1S_0 sequence seem to be essentially unperturbed while the lifetimes of the lower 1D_2 are strongly affected by admixture of $3p^2\ ^1D_2$ character into the wavefunctions. The six lowest $3snd\ ^1D_2$ states together

contain 50 % of the total $3p^2 \ ^1D_2$ wavefunction and the rest is mixed into the higher $3snd \ ^1D_2$ members and the adjacent continuum. Thus, the $3p^2 \ ^1D_2$ state does not exist as a well-defined level and none of the observed levels have been assigned the $3p^2 \ ^1D_2$ symbol¹⁸.

In Table II our experimental lifetimes are compared with lifetimes calculated from theoretical oscillator strengths given by Froese Fischer^{11,12}. Oscillator strengths calculated from the experimental lifetime values using the branching ratio obtained from Refs. 11,12 are also included. In the 1S_0 sequence the agreement between experimental and theoretical lifetimes is excellent. The behaviour of the 1D_2 sequence is also fully explained. The $3p^2 \ ^1D_2$ admixture is strongest in the state designated $3s3d \ ^1D_2$ where the principal quantum numbers all are the same. The oscillator strength for a pure $3p^2 \ ^1D_2 - 3s3p \ ^1P_1$ transition is large but the mixing of $3p^2 \ ^1D_2$ character into the $3snd \ ^1D_2$ wavefunction through interference leads to a reduction of the $\langle \alpha 3snd \ ^1D_2 + \beta 3p^2 \ ^1D_2 | P | 3s3p \ ^1P_1 \rangle$ matrix element for the oscillator strength (P is the dipole operator; $\alpha^2 + \beta^2 = 1$), thus increasing the actual lifetime. This effect is similar to what was recently observed for the $4d5p \ ^1F_3$ perturber in the $5snf \ ^1F_3$ series in Sr¹⁹. The lifetime of a state is in general very sensitive to the relative signs of the interacting wavefunctions. However when calculating the lifetime of a perturbed state high up in a Rydberg sequence it is often reasonable to assume that interference effects average to zero when summing over all the individual transition probabilities. This approach is generally used in MQDT¹⁵. With the assumption above it is then appropriate to only use the square of the mixing coefficients to calculate the amount of perturber character in a state^{20,21}, even if information about the relative signs of the wavefunctions then is lost.

As can be seen the experimental lifetime values support the theoretical calculations by Froese Fischer. Similar theoretical results but with slightly weaker admixture is obtained by Beigang et al. in MQDT calculations based on the positions of energy levels⁶. It would be interesting to further probe the interaction in the $3snd \ ^1D_2$ sequence by studying the hyperfine structure of ^{25}Mg .

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Table I. Experimental values for the natural radiative lifetimes. Excitation wavelength and method of excitation are included. In case of two step excitation, the first step laser was tuned to the $3s^2\ ^1S_0 - 3s3p\ ^1P_1$ transition at $\lambda = 2852.13\ \text{\AA}$.

(ts = two step, tp = two-photon absorption and m = excitation from metastable state)

State	Experimental Lifetimes		Wavelength ^a ($\text{\AA}$)	Methods of Excitation
	This work (ns)	Schaefer ^d (ns)		
$3s4s\ ^1S_0$	47 (3)	163 (8)	4596.00/4621.30	tp, m
$3s5s\ ^1S_0$	100 (5)	201 (4)	3804.30	tp
$3s6s\ ^1S_0$	211 (12)		4730.03	ts
$3s7s\ ^1S_0$	350 (16)		4354.53	ts
$3s8s\ ^1S_0$	548 (35)		4165.10	ts
$3s9s\ ^1S_0$	846 (40)		4054.69	ts
$3s10s\ ^1S_0$	1155 (58)		3984.21	ts
$3s11s\ ^1S_0$	1650 (140)		3936.30 ^b	ts
$3s12s\ ^1S_0$	2150 (180)		3902.15 ^b	ts
$3s13s\ ^1S_0$	2507 (225)		3876.92 ^b	ts
$3s14s\ ^1S_0$	2960 (210)		3857.73 ^b	ts
$3s15s\ ^1S_0$	3500 (260)		3842.79 ^b	ts

Table I continued

3s3d 1D_2	81(6)	57.0(3.6)	4308.76	tp
3s4d 1D_2	57(3)	54.9(1.4)	3762.89	tp
3s5d 1D_2	50(4)	44.3(2.4)	4702.99	ts
3s6d 1D_2	54(3)	50.3(2.2)	4351.91	ts
3s7d 1D_2	70(6)	73.1(2.7)	4167.27	ts
3s8d 1D_2	93(7)	85.4(10.5)	4057.51	ts
3s9d 1D_2	127(8)	99.4(9.4)	3986.75	ts
3s10d 1D_2	167(17)	66(15)	3938.40	ts
3s11d 1D_2	239(25)	37.1(3.4)	3903.86	ts
3s12d 1D_2	298(20)	32.1(2.9)	3978.31	ts
3s13d 1D_2	359(26)		3858.86	ts
3s14d 1D_2	430(30)		3843.63 ^C	ts
3s15d 1D_2	535(50)		3832.76 ^C	ts

a) When nothing else is noted wavelengths are taken from Ref. 22.

b) Calculated values assuming a constant quantum defect of 1.524 in the 1S_0 sequence.

c) Calculated from Ref. 7.

d) From Ref. 8.

Table II. Comparison between the experimental lifetimes and lifetimes calculated from theoretical oscillator-strengths.

State	Lifetimes (ns)		Experimental gf-values ^b in transitions to 3s3p ¹ P ₁
	Experiment	Theory ^a	
3s4s ¹ S ₀	47(3)	45	0.45(3)
5	100(5)	102	0.019(1)
6	211(12)	202	0.0040(2)
3s7s ¹ S ₀	350(16)	362	0.00156(7)
3s3d ¹ D ₂	81(6)	93	0.72(5)
4	57(3)	40	0.49(3)
5	50(4)	38	0.30(3)
6	54(3)	50	0.25(2)
7	70(6)	65	0.17(2)
3s8d ¹ D ₂	93(7)	93	0.13(1)

a) The oscillator strengths for the theoretical values are taken from Refs. 11,12 .

b) Branching ratios taken from Refs. 11,12 have been used.

Figure Captions

Fig. 1 Experimental set-up.

Fig. 2 Schematic energy level diagram for Mg I.

Fig. 3 Three experimental decay curves shown on a logarithmic scale. The $5s\ ^1S_0$ state was excited by two-photon absorption.

Fig. 4 Our experimental lifetime values versus the effective principal quantum number drawn on a log-log scale. A slope corresponding to a n^{*3} -dependence is included.

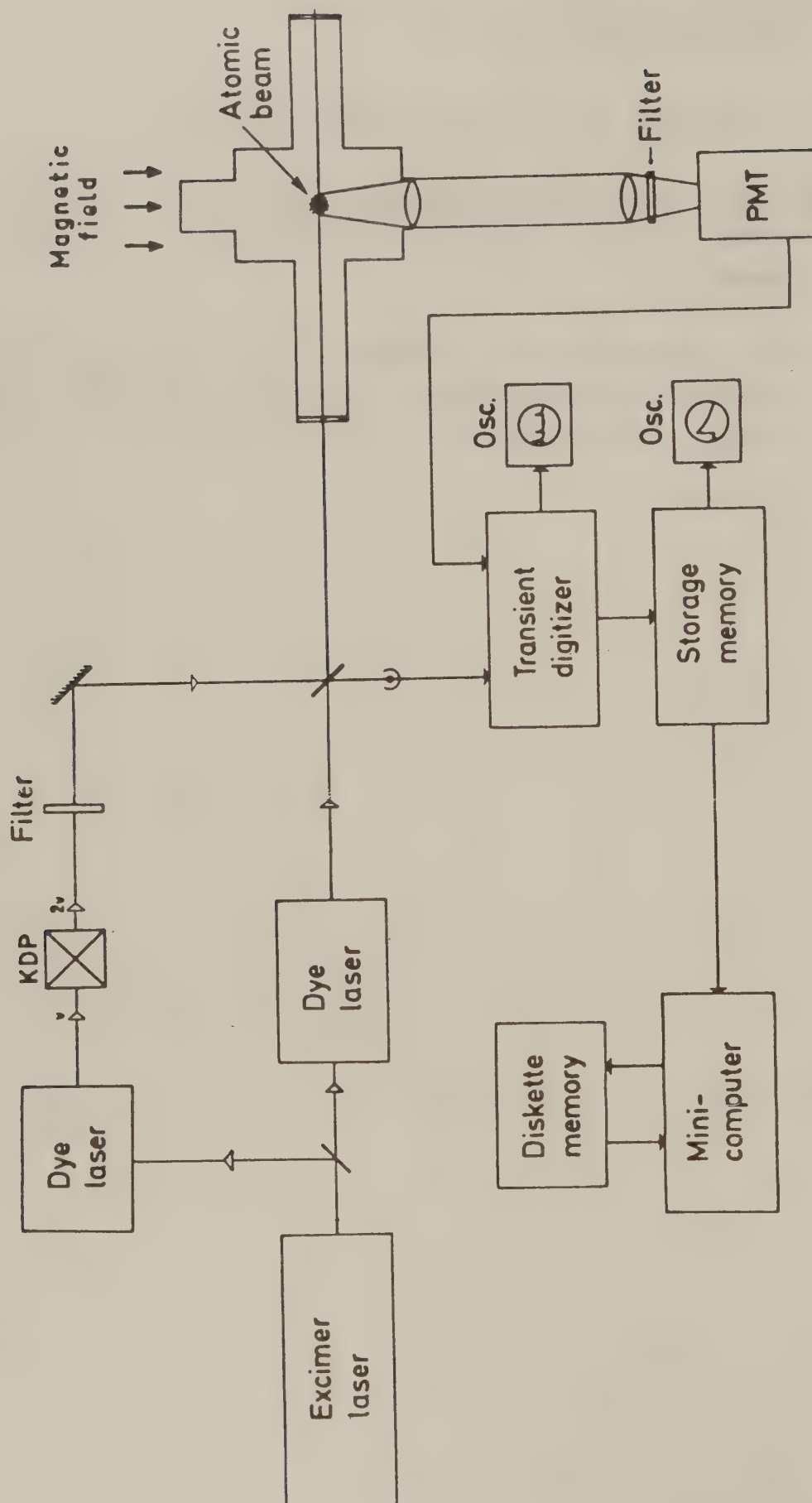


Figure 1.

Mg I

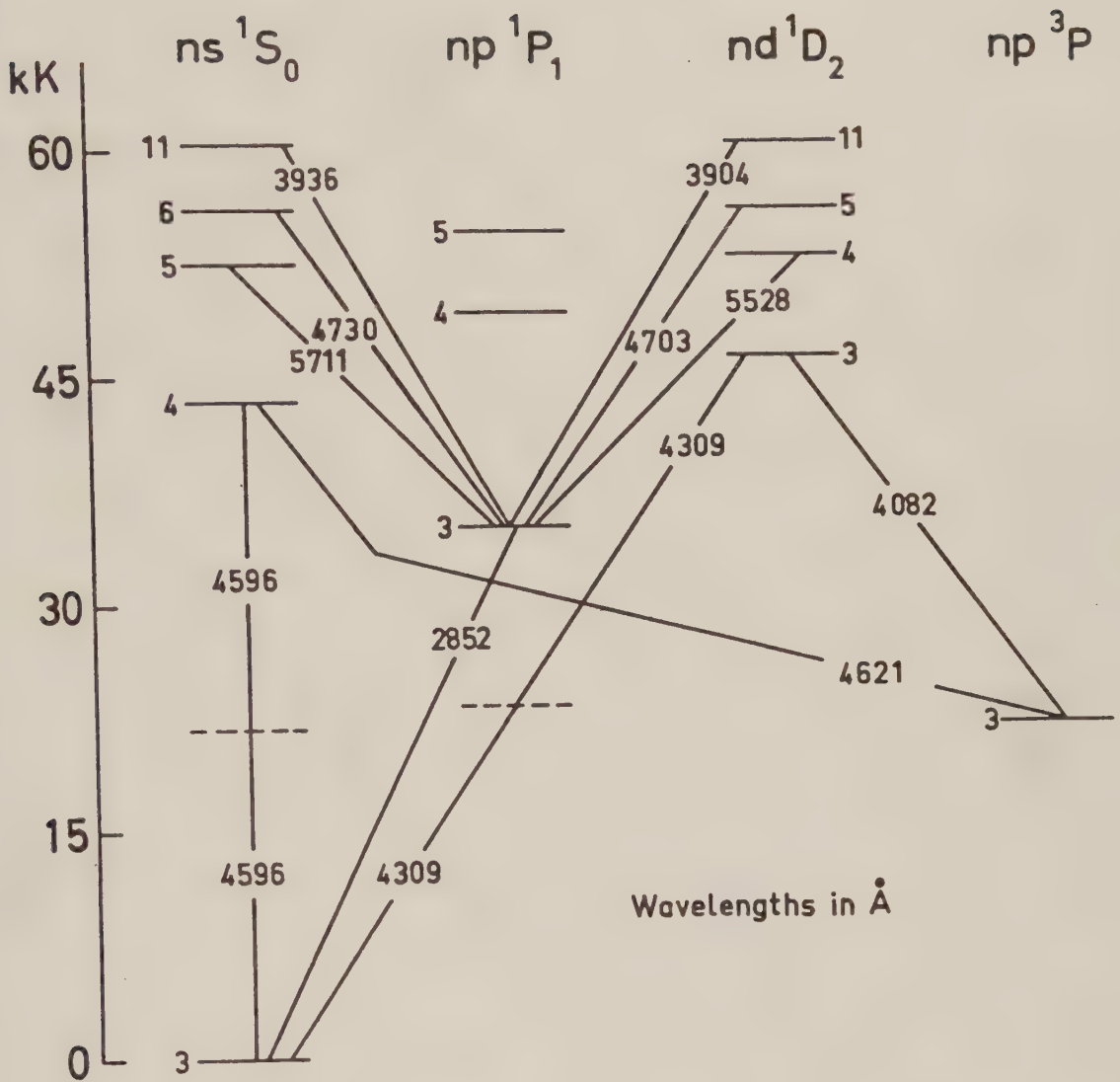


Figure 2.

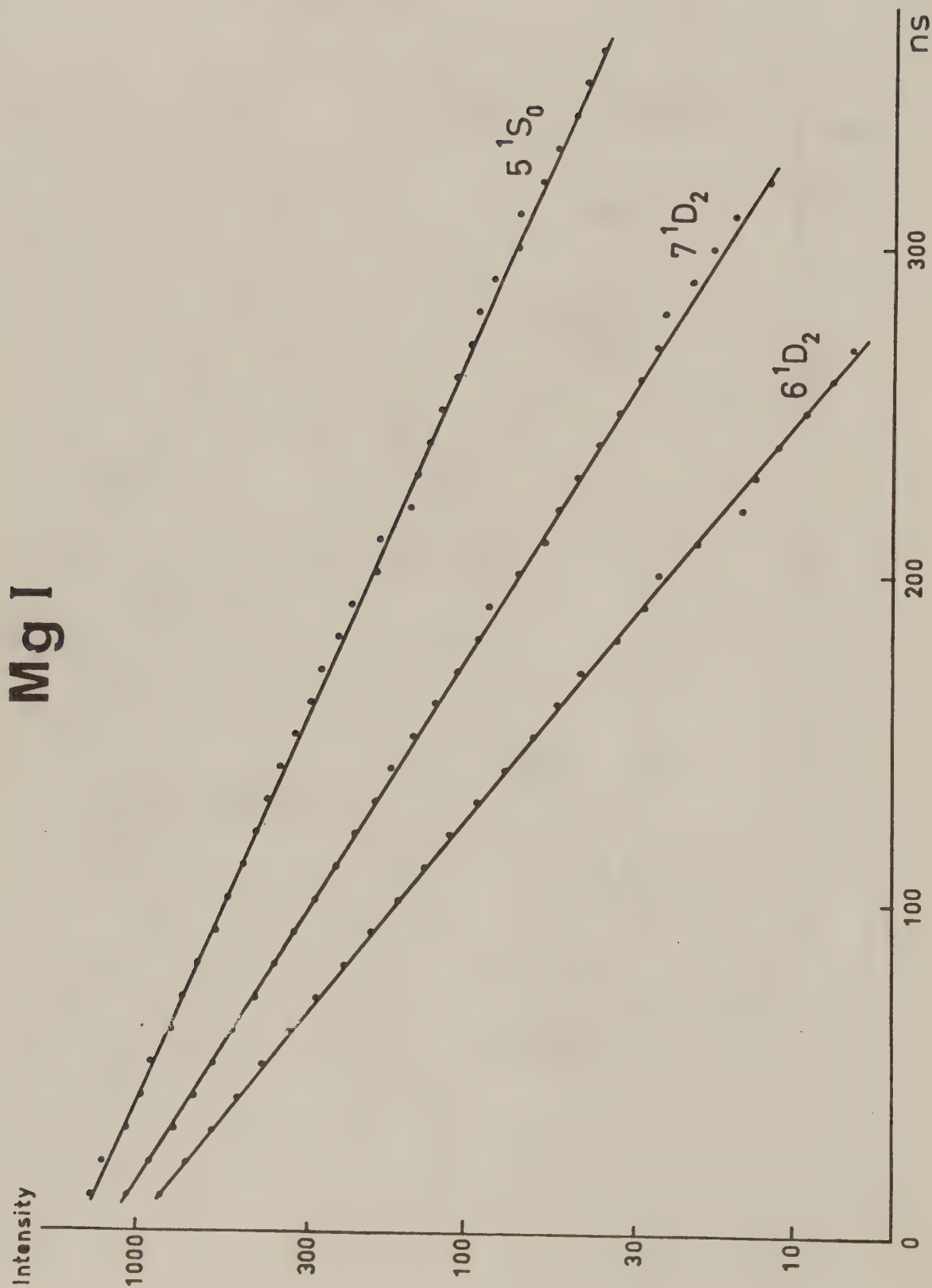


Figure 3.

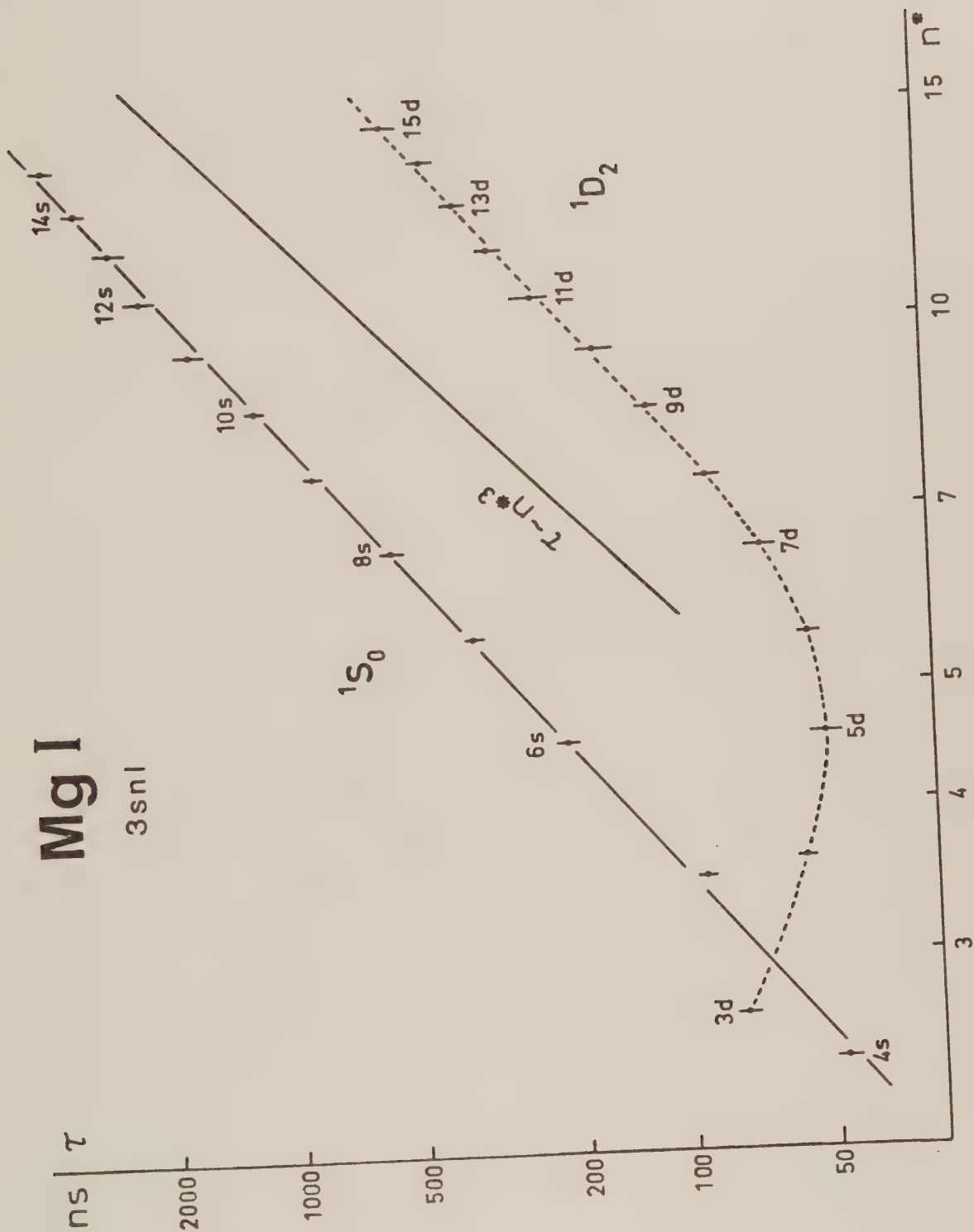


Figure 4.

NATURAL RADIATIVE LIFETIMES AND HYPERFINE STRUCTURE OF THE $4s^2 6p\ ^2P_{3/2,1/2}$ LEVELS OF GALLIUM

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Natural radiative lifetimes and the signs of previously determined hyperfine coupling constants were determined for the $4s^2 6p\ ^2P_{3/2,1/2}$ levels of gallium. The lifetimes were found to be 167(4) and 172(9) ns, respectively. In addition, the quadrupole moments of ^{69}Ga [0.17(3) b] and ^{71}Ga [0.10(2) b] were deduced, and the isotope shifts in the 639.7 and 641.3 nm Ga lines were determined.

Gallium is a group-three element with ground configuration $4s^2 4p$. On excitation of the outer 4p electron, an alkali-like spectrum is produced. However, the ground state is a $^2P_{1/2}$ state. The hyperfine coupling constants of this state and its metastable fine-structure partner $4p\ ^2P_{3/2}$ were measured with high precision using atomic-beam magnetic-resonance spectroscopy [1]. In a theoretical analysis by Bessis et al. [2], the importance of polarization and relativistic effects are shown. Comparatively few excited states have been investigated in gallium. The hfs of the first excited $^2S_{1/2}$ state, $5s$, was recently determined by Neijzen and Dönszelmann [3]. These authors have also performed a quantum-beat experiment on the $6p\ ^2P_{3/2,1/2}$ states [4], yielding the magnetic-dipole and the electric quadrupole coupling constants a and b of the two stable spin 3/2 isotopes ^{69}Ga (60%) and ^{71}Ga (40%) with high precision. However, the signs of the interaction constants cannot be determined in such experiments. In the present paper we report the results of a high-resolution study of the 639.7 nm ($5s\ ^2S_{1/2} - 6p\ ^2P_{3/2}$) and 641.3 nm ($5s\ ^2S_{1/2} - 6p\ ^2P_{1/2}$) gallium lines yielding these signs and, in addition, the optical isotope shifts in these lines. A multi-mode and a single-mode dye laser were used for step-wise excitation of a collimated atomic beam. By pulse-modulating the second-step laser acousto-optically, the natural lifetimes of the P states, previously known only with low precision [4], were determined in delayed-coincidence experiments.

The experimental set-up used in the high-resolution experiments is similar to the one used in recent investigations on In [5] and Al [6] in our laboratory. The principles for the special kind of delayed-coincidence we used (PUMOLS) can be found in ref. [7] and the experimental arrangement is similar to the one used in ref. [8].

In fig. 1 a recording of the 641.3 nm line is shown together with an excitation scheme and a hyperfine-transition diagram. From the relative intensities of the hyperfine components it is evident that the a factors are positive in the $^2P_{1/2}$ and the $^2S_{1/2}$ states for both isotopes. However, because of line broadening due to the short-lived intermediate S-state ($\tau \approx 6$ ns [9]) and residual Doppler effect, the deduced a factors are less precise than, although consistent with, the results given by Neijzen and Dönszelmann ($|a_{1/2}(71)| = 67.4(7)$ MHz; $|a_{1/2}(69)| = 53.3(7)$ MHz; $|a_{5s}(71)| = 2716.0(30)$ MHz; $|a_{5s}(69)| = 2137.7(20)$ MHz for ^{69}Ga and ^{71}Ga) [3,4].

For the even less resolved 639.7 nm line, a computer program, generating a resulting overlapped structure from a given set of interaction constants and a chosen experimental linewidth, was used for establishing that the a and b factors for the $6p\ ^2P_{3/2}$ state given in ref. [4] ($|a_{3/2}(71)| = 28.5(2)$ MHz; $|b_{3/2}(71)| = 2.4(3)$ MHz; $|a_{3/2}(69)| = 22.4(2)$ MHz; $|b_{3/2}(69)| = 3.2(3)$ MHz) are positive for both isotopes. (The quantum-beat experiment establishes that the a and b factors have the same signs.) From a large number of experi-

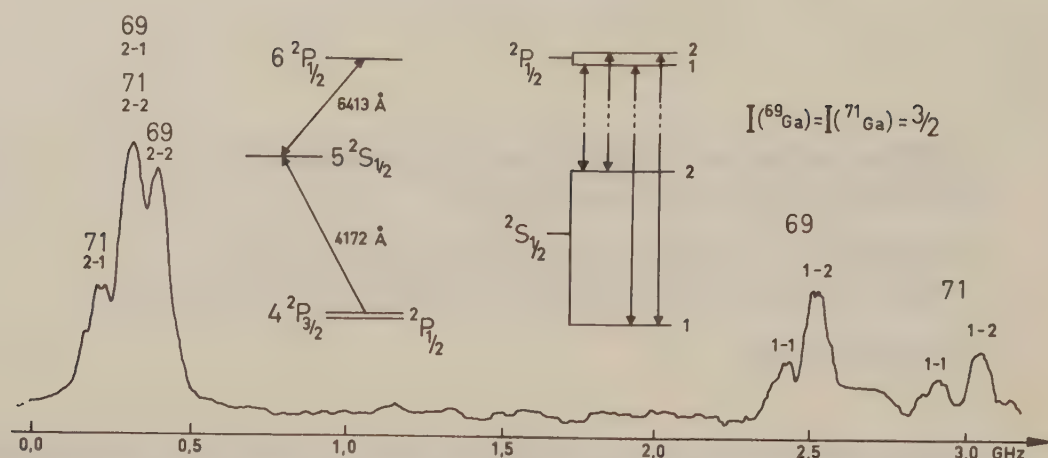


Fig. 1. Experimental recording of the 641.3 nm Ga line ($5^2S_{1/2} - 6^2P_{1/2}$) and diagrams illustrating the step-wise excitation scheme and hyperfine transitions.

mental curves, the isotope shifts $\Delta\nu(71-69)$ were also determined, employing the coupling-constant results of refs. [3,4]. We find $\Delta\nu(71-69) = 149(8)$ MHz for the 641.3 nm line and $\Delta\nu(71-69) = 143(8)$ MHz for the 639.7 nm line. The error bars are determined by the experimental scattering and the estimated uncertainty in the used deconvolution procedure. The cor-

responding normal mass shifts (nms) are 105 MHz for both lines. The specific mass shift (sms) for an S-P transition is estimated by Heilig and Steudel [10] as $\Delta\nu(\text{sms}) = (0.3 \pm 0.9) \Delta\nu(\text{nms})$ or 31 ± 94 MHz in our case. This leaves 13 and 7 MHz with large uncertainties for the field shift of the 641.3 and 639.7 nm lines, respectively.

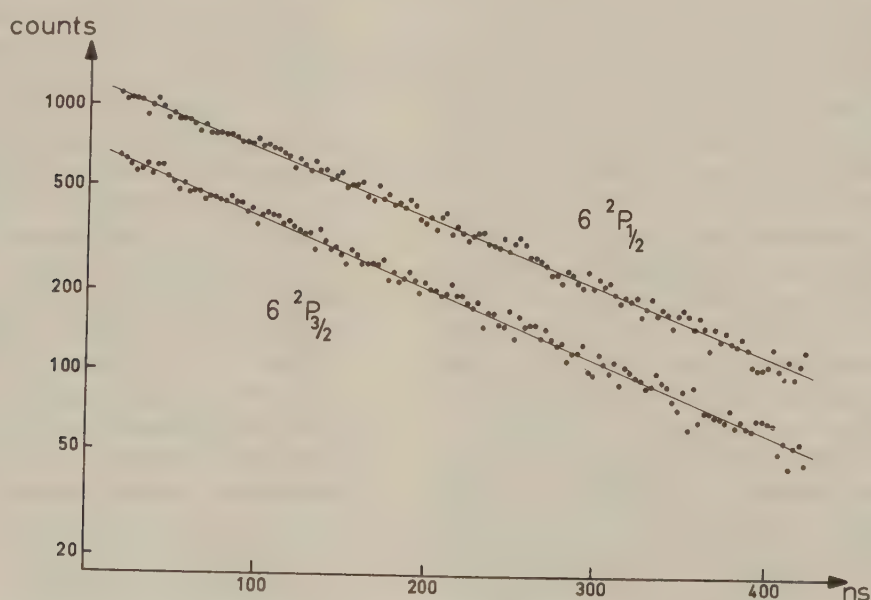


Fig. 2. Experimental decay curves shown on a logarithmic scale. The background (about 50 counts) has been subtracted. Exponential fits to the data are included.

Table 1

Results for ^{69}Ga . One-electron expressions from ref. [13] have been used with the following parameters: $F_F(1/2) = 1.0757$, $F_F(3/2) = 1.0154$, $H_F = 1.0174$, $R_F = 1.0314$, $Z_i = 27$. Experimental values are from refs. [1,4].

State	$a_{3/2}$ (MHz)	$a_{3/2,\text{corr}}$ (MHz)	$\langle a_0^3/r^3 \rangle$ from $a_{3/2,\text{corr}}$	$\langle a_0^3/r^3 \rangle$ from δW	b (MHz)	Q (barn)
$6p^2P_{3/2}$	22.4(2)	12.0	0.164	0.171	3.2(3)	0.197
$4p^2P_{3/2}$	190.79428(15)	242.9	3.33	3.43	62.52247(30)	0.191

In our lifetime measurements the red laser was pulse-modulated acousto-optically. A sufficiently high magnetic field was applied to prevent Zeeman quantum beats due to the earth's magnetic field to influence the results. In fig. 2 examples of experimental curves with fitted exponentials are shown on a logarithmic scale. We obtain $\tau(6p^2P_{1/2}) = 172(9)$ ns and $\tau(6p^2P_{3/2}) = 167(4)$ ns in agreement with ref. [4] [160(35) ns and 150(15) ns, respectively], but considerably more precise. Our error bars are determined from the statistical scattering of data and with consideration of possible systematic effects.

We will finish this paper with a simple analysis of the discussed hyperfine structure, based on the assumption that the radial parameters for the orbital and the spin-dipole interactions are equal. The procedure reviewed in ref. [11] and applied for alkali-atom P states, e.g. in ref. [12] was shown to apply remarkably well to In P states [5]. In this semi-empirical approach, the effect of core polarization a_c can be isolated and corrected a factors, referring to the p-electron alone can be calculated. In table 1 the results of this calculation of the radial parameters for the 4p and 6p states are given for ^{69}Ga together with values derived from the observed fine structure δW using simple one-electron expressions [13]. It can be seen that the core polarization is quite significant and of different sign for the 4p and 6p states. Quite consistent results are obtained from the fine and hyperfine structure. Using the mean value for the radial parameter together with the experimental b factors, the quadrupole moment Q for ^{69}Ga is also calculated to be 0.197 b and 0.191 b from the 6p and 4p states, respectively. Corresponding Q -values for ^{71}Ga are 0.117 b and 0.120 b. The values are remarkably consistent. Using the Sternheimer correcting factor 0.886 [14], calculated for the 4p state also for the 6p state, we obtain as "best" values for the quadrupole

moments $Q(^{69}\text{Ga}) = 0.17(3)$ b and $Q(^{71}\text{Ga}) = 0.10(2)$ b. Here the error bars are mainly determined by the estimated uncertainty in the used evaluation procedure. For obtaining more accurate results, a many-body calculation [15] should be performed.

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Lifetime measurements in the $^2S_{1/2}$ and $^2D_{3/2,5/2}$ sequences of indium

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Natural radiative lifetimes in the $5s^2ns\ ^2S_{1/2}$ and $5s^2nd\ ^2D_{3/2,5/2}$ ($n \leq 20$) sequences of indium have been measured using pulsed laser excitation of an atomic beam. The uv pulses were produced by a YAG-pumped, or alternatively, an excimer-pumped dye-laser system. While the S -state lifetimes show a normal $(n^*)^3$ behavior, the D sequences are strongly perturbed. The lifetimes of the $5s\ 5p^2\ ^4P$ states were found to be J dependent.

I. INTRODUCTION

During the last few years, highly excited atomic states have been extensively studied regarding radiative properties as well as energy substructure. Using laser-spectroscopy techniques, especially, the alkali-metal and alkaline-earth elements have been investigated. In this paper we report on lifetime measurements on the group-III element In. Indium has three electrons outside closed shells but two electrons usually form a closed subshell. The basic spectrum is then alkali-metal-like with a 2P state as ground level. The group-III atoms are well suited for laser-spectroscopic investigations since the 2S and 2D sequences are directly accessible from the ground configuration in one-step excitation procedures provided short uv laser-wavelengths can be obtained. In our measurements, lifetimes in the $5s^2ns\ ^2S_{1/2}$ and $5s^2nd\ ^2D_{3/2,5/2}$ sequences up to $n=20$ were determined. Two different laser systems were used alternatively; a Nd:YAG-pumped Rhodamine dye laser employing frequency doubling and anti-Stokes Raman shifting, or an excimer-pumped Coumarine-47 dye laser employing frequency doubling.

Although alkali-metal-like, the In spectrum is complicated by the presence of states with two excited electron orbitals. The $5s5p^2$ configuration has states both above and below the ionization limit and can cause perturbations in the Rydberg sequences. Such perturbations have been studied very extensively in the alkaline-earth elements and influence lifetime values¹ as well as g_J factors,² hyperfine structures, and isotope shifts.^{3,4}

The basic energy-level structure in In has been determined by Garton and Codling in absorption measurements.⁵ Recently the investigations were extended to high principal quantum numbers using laser-spectroscopy methods.⁶ The energy levels were

found to be affected by perturbations. Only a few hyperfine structures have been measured so far. The hyperfine structure in the lowest 2D states, measured by the level-crossing technique, was found influenced by interaction with the $5s\ 5p^2$ configuration,^{7,8} while the hyperfine structure in the lowest 2P states is affected by core polarization effects.⁹ Very recently the lifetimes of the lowest members of the 2S and 2D sequences have been measured.¹⁰

II. EXPERIMENTAL TECHNIQUE

The setup used in the present investigation is shown in Fig. 1. During the measurements we used one out of the two laser systems shown. The frequency-doubled green light from a Nd:YAG-laser (Quanta Ray DCR-1A) was used for pumping a dye laser (Quanta Ray PDL-1) with Rhodamine-590, Rhodamine-640, or Kiton red dye. The 532-nm pump pulses had an energy of about 300 mJ. The dye laser light was frequency doubled into the uv region using a KD*P (deuterated potassium dihydro-

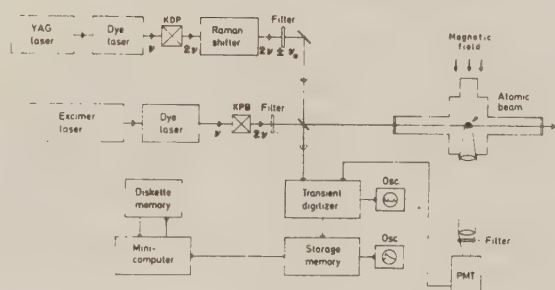


FIG. 1. Experimental setup showing the two laser systems used alternatively.

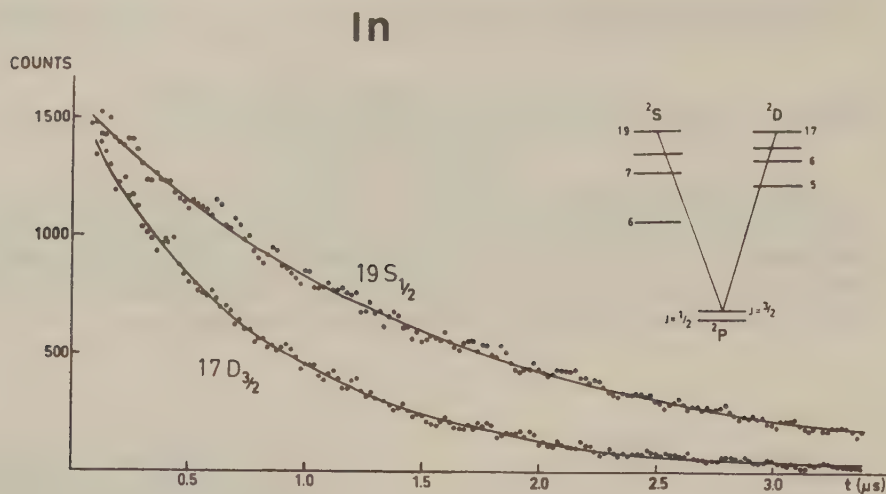


FIG. 2. Experimental data points in arbitrary units and fitted exponentials for $19^2S_{1/2}$ and $17^2D_{3/2}$. Data were sampled for 2000 laser shots. A schematic energy-level diagram is included.

gen phosphate) crystal. The uv light was passed through a Raman-shifter (Quanta Ray RS-1) operating with 10 atm of H_2 gas. Every Raman shift changes the photon energy by 4155 cm^{-1} . Most of the light passes through unshifted, but the Stokes components decrease energy and the anti-Stokes components increase energy. With a suitable laser dye, frequency doubling, and combination with up to second-order anti-Stokes shifting, we could produce tunable light for all the studied transitions 227–451 nm. The excitation energy varied between 0.01 and 20 mJ depending on the laser dye and the nonlinear process. In some cases we used a mixture between Rhodamine-610 and Rhodamine-590 dyes to optimize the power in critical wavelength ranges. The major part of the measurements were made with the Nd:YAG-laser system and all the lifetimes were measured at least at two different occasions using this system.

In some of the measurements we used an excimer laser (Lambda Physik EMG 102) with XeCl gas mixture pumping a dye laser (Lambda Physik FL 2002) with Coumarine-47 dye. The dye laser light was frequency doubled with a KPB ($KB_5O_8 \cdot 4H_2O$) crystal. The 308-nm pump pulses had an energy of about 150 mJ using 10 Hz repetition rate. The tunable uv pulses had an energy of approximately 0.01 mJ at 230 nm. The smaller pulse energy generated by the excimer laser system was to some extent compensated by a narrower spectral width.

An atomic beam of In was created in a high-vacuum system. The indium atoms were excited from the $^2P_{1/2}$ ground or the $^2P_{3/2}$ thermally populated metastable state to the $^2S_{1/2}$ and $^2D_{3/2,5/2}$ states. The exciting light was passed in and out of the vacuum system through long tubes in order to reduce stray light. The fluorescent light from the excited states was analyzed using narrow interfer-

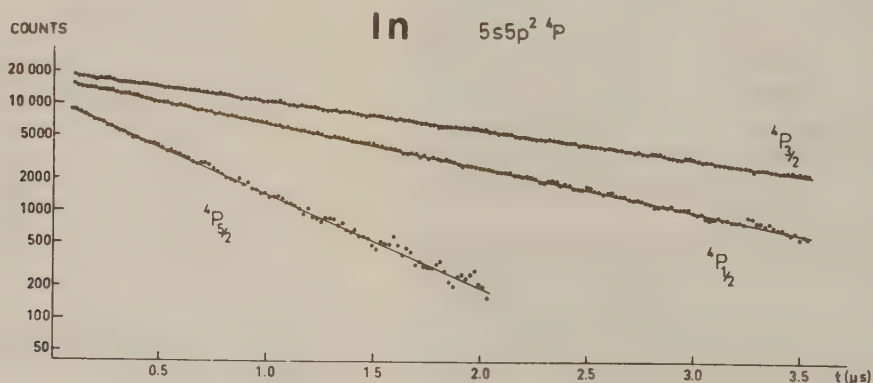


FIG. 3. Logarithms of the decay curves for the three states of the $5s5p^2 4P$ term.

TABLE I. Experimental values for natural radiative lifetimes. Excitation wavelengths and the methods of wavelength generation are included. Normally the excitation starts from the $^2P_{3/2}$ metastable level.

State	Lifetime (ns)		Wavelength (Å)	Specifics of Nd:Yag-pumped dye-laser excitation	Additional measurements with excimer system (X)
	this work	other works			
$5s^2 6s^2 \ ^2S_{1/2}$		7.0(3) ^b	4511.31		
$5s^2 7s^2 \ ^2S_{1/2}$	27(6)	19.5(15) ^c	2753.88 ^a	R590,D	
$5s^2 8s^2 \ ^2S_{1/2}$	55(6)	53(5) ^c	2601.75	KR,D,1AS	
$5s^2 9s^2 \ ^2S_{1/2}$	104(12)	118(10) ^c	2468.02	R590,D,1AS	X
$5s^2 10s^2 \ ^2S_{1/2}$	163(13)	200(20) ^c	2278.20 ^a	R590,D,2AS	
$5s^2 11s^2 \ ^2S_{1/2}$	244(19)		2358.70	KR,D,2AS	
$5s^2 12s^2 \ ^2S_{1/2}$	330(21)		2332.76	KR,D,2AS	X
$5s^2 13s^2 \ ^2S_{1/2}$	490(35)		2315.09	R590 + R610,D,2AS	X
$5s^2 14s^2 \ ^2S_{1/2}$	625(60)		2302.49	R590,D,2AS	X
$5s^2 15s^2 \ ^2S_{1/2}$	785(70)		2293.17	R590,D,2AS	X
$5s^2 16s^2 \ ^2S_{1/2}$	1025(70)		2286.09	R590,D,2AS	X
$5s^2 17s^2 \ ^2S_{1/2}$	1170(95)		2280.59	R590,D,2AS	
$5s^2 18s^2 \ ^2S_{1/2}$	1360(135)		2276.21	R590,D,2AS	
$5s^2 19s^2 \ ^2S_{1/2}$	1690(200)		2272.67	R590,D,2AS	
$5s^2 20s^2 \ ^2S_{1/2}$	2000(300)		2269.80	R590,D,2AS	
$5s^2 5d^2 \ ^2D_{3/2}$		7.0(4) ^d	3258.56		
$5s^2 6d^2 \ ^2D_{3/2}$		25.5(10) ^d	2713.94		
$5s^2 7d^2 \ ^2D_{3/2}$	200(14)		2522.98	R590,D,1AS	
$5s^2 8d^2 \ ^2D_{3/2}$	317(22)		2430.99	R640,D,2AS	X
$5s^2 9d^2 \ ^2D_{3/2}$	550(50)		2379.00	KR,D,2AS	X
$5s^2 10d^2 \ ^2D_{3/2}$	455(40)		2346.56	KR,D,2AS	X
$5s^2 11d^2 \ ^2D_{3/2}$	490(50)		2324.92	KR,D,2AS	X
$5s^2 12d^2 \ ^2D_{3/2}$	485(40)		2309.75	R590,D,2AS	X
$5s^2 13d^2 \ ^2D_{3/2}$	500(30)		2298.70	R590,D,2AS	
$5s^2 14d^2 \ ^2D_{3/2}$	570(40)		2290.42	R590,D,2AS	
$5s^2 15d^2 \ ^2D_{3/2}$	635(40)		2283.98	R590,D,2AS	
$5s^2 16d^2 \ ^2D_{3/2}$	735(60)		2278.96	R590,D,2AS	
$5s^2 17d^2 \ ^2D_{3/2}$	820(65)		2274.94	R590,D,2AS	
$5s^2 18d^2 \ ^2D_{3/2}$	895(60)		2271.65	R590,D,2AS	
$5s^2 19d^2 \ ^2D_{3/2}$	1075(70)		2268.94	R590,D,2AS	
$5s^2 20d^2 \ ^2D_{3/2}$	1275(115)		2266.70	R590,D,2AS	

ence filters and detected with a photomultiplier tube (EMI 9558 BQ). Both excitation and detection were at right angles to the atomic beam.

After every laser pulse the fluorescence decay was captured by a transient digitizer (Biomation 8100). The transient digitizer was triggered by a photodiode detecting a small fraction of the dye-laser light. Every channel in the transient digitizer corresponds to 10 ns. The time scale was calibrated using a radio frequency signal. The digitized data was transferred to a storage memory where a large number of transients could be added. In order to get a good signal-to-noise ratio about 1000 transients were added before the signal was transferred to a mini-computer (Digital Equipment Corporation PDP-11V03) and stored on a floppy disc.

III. MEASUREMENTS AND RESULTS

The excitation wavelengths for reaching the individual states were calculated from the data given in Refs. 11 and 5. The actual wavelength setting for the dye laser was calculated considering the frequency doubling and Raman shifting. To avoid the influence of slow Zeeman quantum beats, a set of Helmholtz coils supplied a sufficient magnetic field in the excitation volume to cause beats far beyond the high-frequency out-off of the detection system. To reduce radio frequency disturbances originating from the pump laser (especially the excimer laser) we used a very short cable between the photomultiplier tube and the *A* input of the transient digitizer. With a pickup cable of suitable length in the *B* input

TABLE I. (Continued.)

State	Lifetime (ns)		Wavelength (Å)	Specifics of Nd:Yag-pumped dye-laser excitation	Additional measurements with excimer system (X)
	this work	other works			
$5s^25d^2D_{5/2}$		7.1(6) ^c	3256.09		
$5s^26d^2D_{5/2}$		18.6(15) ^c	2710.27		
$5s^27d^2D_{5/2}$	147(10)	154(10) ^c	2521.37	R590,D,1AS	
$5s^28d^2D_{5/2}$	238(20)	300(60) ^c	2429.86	R640,D,2AS	X
$5s^29d^2D_{5/2}$	640(50)		2378.14	KR,D,2AS	X
$5s^210d^2D_{5/2}$	780(80)		2345.90	KR,D,2AS	X
$5s^211d^2D_{5/2}$	760(80)		2324.41	KR,D,2AS	X
$5s^212d^2D_{5/2}$	780(65)		2309.32	R590,D,2AS	X
$5s^213d^2D_{5/2}$	845(60)		2298.33	R590,D,2AS	X
$5s^214d^2D_{5/2}$	1005(70)		2290.10	R590,D,2AS	X
$5s^215d^2D_{5/2}$	1185(110)		2283.75	R590,D,2AS	X
$5s^216d^2D_{5/2}$	1335(125)		2278.80	R590,D,2AS	X
$5s^217d^2D_{5/2}$	1540(200)		2274.79	R590,D,2AS	
$5s^218d^2D_{5/2}$	1810(210)		2271.55	R590,D,2AS	X
$5s^219d^2D_{5/2}$	1985(250)		2268.87	R590,D,2AS	
$5s^220d^2D_{5/2}$	2215(250)		2266.65	R590,D,2AS	
$5s5p^2^4P_{1/2}$	1065(65)		2858.13 ^a	R590,D	
$5s5p^2^4P_{3/2}$	1645(110)		2775.36 ^a	R590,D	
$5s5p^2^4P_{5/2}$	510(30)		2836.70	R590,D	

^aExcitation from the ground level $^2P_{1/2}$; R, Rhodamine; KR, Kiton Red; D, frequency doubling; 1AS, 1 anti-Stokes shift; 2AS, 2 anti-Stokes shifts.

^bReference 14.

^cReference 10.

^dReference 7.

^eReference 8.

of the transient digitizer we could eliminate the radio frequency disturbances using an *A*-minus-*B* option.

When a decay curve had been transferred to the minicomputer, a program was used for obtaining the best exponential fit to the experimental data. On some short-lived states we recorded both the fluorescent light and the exciting laser pulse. From the laser pulse shape, a computer program generated decay curves for different lifetimes to make the best fit to our experimental decay curve. Figures 2 and 3 show the quality of some experimental curves. Background counts are subtracted and the fitted exponentials are included.

For each state, recordings were made on several occasions and some states were measured with both laser systems. Special attention was paid to ensure that there was no influence due to saturation in the photomultiplier tube, multiple scattering, collisions, Zeeman quantum beats, and flight-out-of-view effects.¹² Table I gives our experimentally determined lifetimes for the $^2S_{1/2}$ and $^2D_{3/2,5/2}$ Rydberg sequences and the doubly excited configuration

$5s5p^2^4P$. The table includes excitation wavelengths, wavelength-generating system, and lifetime values measured by other groups. The error bars in our measurements include both statistical scattering and estimated possible systematic error effects in the used method. No correction has been applied for transitions induced by blackbody radiation (see, e.g., Ref. 13). Thus the given values pertain to room temperature. The blackbody correction is probably of the order of a few percent for the most long-lived states.

IV. DISCUSSION

The results from the lifetime measurements are plotted in Figs. 4 and 5. For the lower states values are taken from Refs. 14, 7, 8, and 10. In an unperturbed Rydberg sequence the lifetimes are expected to be proportional to a power, close to three, of the effective principal quantum number. For the $^2S_{1/2}$ states, as can be seen in the log-log scale of Fig. 4, the values fall on a straight line, which demonstrates the unperturbed character of this sequence. From a

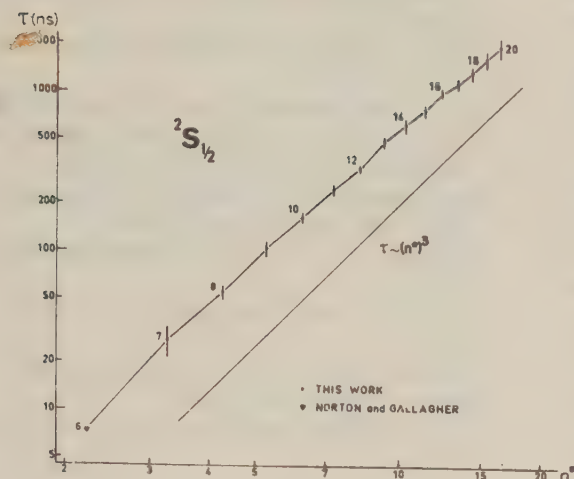


FIG. 4. Experimental lifetimes for the $5s^2ns^2S_{1/2}$ ($n=6-20$) states plotted vs the effective principal quantum number. A straight line showing the lifetime trend in the case of an $(n^*)^3$ dependence is also drawn. Our error bars are represented by the vertical lines.

fit we obtain $\tau = 0.955 n_{\text{eff}}^{2.77}$. The 2D sequences, however, show several strange features. For both the $^2D_{3/2}$ and $^2D_{5/2}$ states there is a drastic increase in lifetime values around $n=7-10$. There is also a large difference in lifetimes between the two fine-structure levels. For low n values the $^2D_{5/2}$ states have a shorter lifetime than the $^2D_{3/2}$ states, while for high n values the situation is reversed.

The effect of perturbations on lifetime values have been studied in several Rydberg sequences in the alkaline-earth elements (Ref. 1 and references therein). In these sequences short-lived states with two excited electrons mix into the energetically close-lying Rydberg levels and cause a decrease in the lifetimes. This effect can be used to determine the degree of configuration mixing. In the 2D sequences of indium, however, the situation is more complicated since no doubly-excited states exist in the energy range of the $(7-10)^2D$ states. In the measurements, the fluorescent light obtained from the atomic beam was one or two orders of magnitude weaker for these states than for the next higher levels. Low-transition probabilities have also been found in absorption measurements⁵ and a similar behavior persists in Ga and Tl. The low-absorption cross sections for these states in In have been sug-

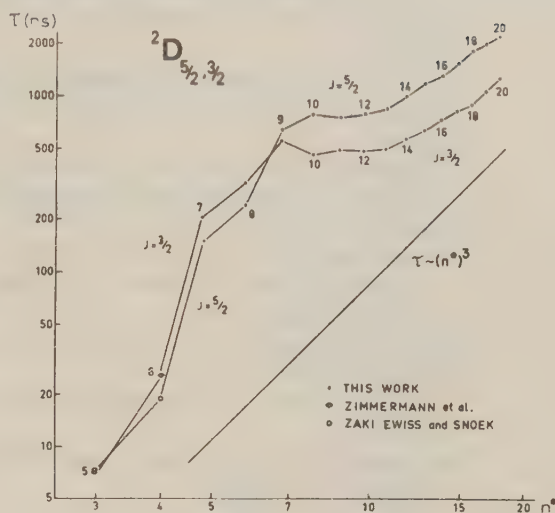


FIG. 5. Experimental lifetimes for the $5s^2nd^2D_{3/2,5/2}$ ($n=5-20$) states plotted vs the effective principal quantum number. Experimental results from earlier measurements are taken from Refs. 7, 8, and 10.

gested to be owing to configuration interaction with the autoionizing $5s5p^2^2D$ states.¹⁵ As can be seen in Fig. 5, the lifetimes increase monotonically for higher states but the lifetime values are different in the $^2D_{5/2}$ and $^2D_{3/2}$ sequences. This indicates that a perturbation strongly affects the levels around $n=7-10$ and in a more smooth way the rest of one or both sequences. Clearly, theoretical calculations including configuration-interaction effects are necessary to explain these results.

Lifetimes for the three $5s5p^2^4P$ states were also measured. In pure LS coupling these states would be metastable and the lifetime values are probably a measure of the degree of intermediate coupling. From a fit of energy levels no deviation from LS coupling is obtained, but for similar elements, e.g., Tl I, and Sn II, and $J = \frac{3}{2}$ have the highest purity and $J = \frac{5}{2}$ the lowest,¹⁶ as indicated by our lifetime results.

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Natural Radiative Lifetimes in the $^2S_{1/2}$ and $^2D_{5/2,3/2}$ Sequences of Aluminum

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Natural radiative lifetimes have been measured for the $3s^2ns\ ^2S_{1/2}$ ($n \leq 10$) and $3s^2nd\ ^2D_{5/2,3/2}$ ($n \leq 11$) sequences of aluminum using pulsed laser excitation of an atomic beam. The investigated states were populated from the ground configuration using UV pulses of wavelength down to 210 nm produced by a Nd:YAG- or an excimer-pumped dye-laser system. The S -state lifetimes increase regularly with increasing principal quantum number but this is not the case for the D sequence.

1. Introduction

In a recent investigation of lifetimes in excited states of indium remarkable irregularities were found for the $^2D_{5/2,3/2}$ sequences [1]. In the present paper we report on lifetime measurements in the lighter group three element aluminum. Being a light element aluminum is particularly suited for theoretical calculations and a considerable effort has been devoted to its radiative properties [2, 3]. The group three elements Al, Ga, In and Tl have three electrons outside closed shells but two electrons usually form a closed subshell. The energy-level structure is then alkali-like but complicated by the presence of states with two excited electron orbitals. Perturbations in Rydberg sequences caused by such doubly-excited states and their effect on lifetime values have been studied very extensively in the alkaline-earth elements (see Ref. 4 and Refs. therein).

Since the ground state in aluminum is a 2P state, the 2S and 2D sequences are accessible by laser excitation in one step provided that short UV wavelengths can be produced. In the measurements of lifetimes in the $3s^2ns\ ^2S_{1/2}$ ($n \leq 10$) and $3s^2nd\ ^2D_{5/2,3/2}$ ($n \leq 11$) sequences two different pulsed laser systems were used alternatively. Using an excimer-pumped coumarin dye-laser and frequency-doubling, wavelengths down to 217 nm was obtained. Laser pulses of shorter wavelength were produced by a Nd:YAG-pumped dye-laser employing frequency-doubling and three anti-Stokes Raman shifts in a

hydrogen high-pressure cell. The lifetime values were determined by time-resolved detection of fluorescent light from an atomic beam.

Wavelengths for the sharp and diffuse series in aluminum have been determined in absorption measurements and a large number of oscillator strengths were obtained employing the hook method [5]. Regarding lifetimes only the lowest 2S and 2D states have earlier been investigated [6, 7].

2. Experimental Techniques

The experimental set-up used in this work is similar to the one described in [1]. About a third of the measurements were performed using an Excimer Laser (Lambda Physik EMG 102) with XeCl gas mixture. The 308 nm excimer light pumped a dye laser (Lambda Physik FL2002) with Coumarin 450 or 470 dye. The dye laser light was frequency-doubled using a KPB crystal. With this laser system wavelengths down to 217 nm were generated.

The major part of the measurements was performed using a Nd:YAG laser (Quanta Ray DCR-1A). The frequency-doubled 532 nm green light pumped a dye laser (Quanta Ray PDL-1) with Rhodamine 590, Rhodamine 640 or Kiton Red dye. The dye laser light was frequency doubled using a KD*P-crystal and was passed through a Raman shifter (Quanta

Ray RS-1) operating with 10 atm. of H_2 gas. Part of the light was then shifted and every Raman-shift changes the photon energy by $4,155\text{ cm}^{-1}$. With a suitable laser dye in combination with frequency doubling and up to third-order anti-Stokes shifting we could produce tunable light down to 210 nm.

An atomic beam of Al was created in a high-vacuum system. The aluminum atoms were excited from the thermally populated metastable $^2P_{3/2}$ state to the $^2S_{1/2}$ - and $^2D_{5/2,3/2}$ -states. The fluorescence light coming from the excited states passed through interference filters and was detected with a photomultiplier tube.

The fluorescence decay was captured by a transient digitizer (Biomation 8100). The digitized data was added in a storage memory until a good signal-to-noise ratio was achieved. After typically one thousand transients the signal was transferred to a floppy disc for later analysis with a mini-computer system (PDP-11V03).

3. Measurements and Results

The excitation wavelengths for the measured states were calculated from data given in [5] and [8]. The dye laser setting had to account for both frequency doubling and Raman shifting. To avoid influence of slow Zeeman quantum beats a sufficiently large magnetic field was applied over the excitation volume. Radiofrequency disturbances originating from the pump laser (especially the excimer laser) were eliminated by subtracting the signal from a pick-up cable.

When a decay curve had been transferred to the mini-computer a program was used to obtain the best exponential fit to experimental data for cases when the lifetimes were longer than 50 ns. Figure 1 shows three typical such experimental recordings. For evaluating shorter lifetimes we also recorded the laser pulse. The dye laser was then set off resonance and a scattering rod was adjusted until the laser-pulse transients had the same height as the previously recorded fluorescence transients. From the laser pulse shape a computer program generated decay curves corresponding to different lifetimes. The computer generated curves were fitted to our experimental fluorescence curve. Figure 2 shows an example of the used procedure.

For each state recordings were made on several occasions and some states were measured with both laser systems. Normal precautions were taken to avoid influence of multiple scattering, collisions, flight-out-of-view effects, Zeeman quantum beats and saturation in the photomultiplier tube [9].

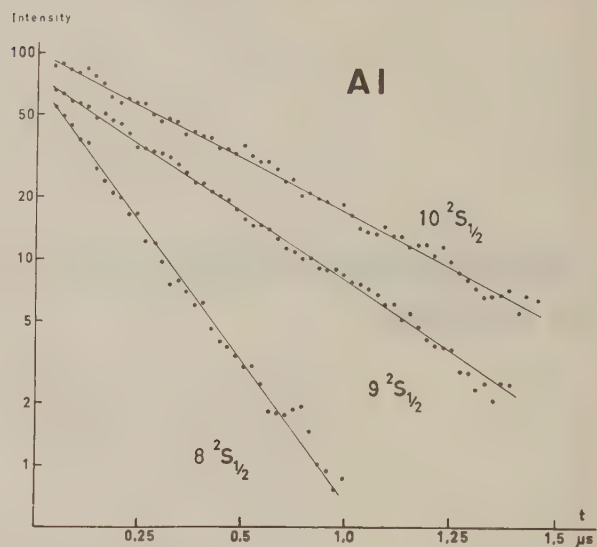


Fig. 1. Experimental decay curves for the 8-10 $^2S_{1/2}$ states shown on a logarithmic scale

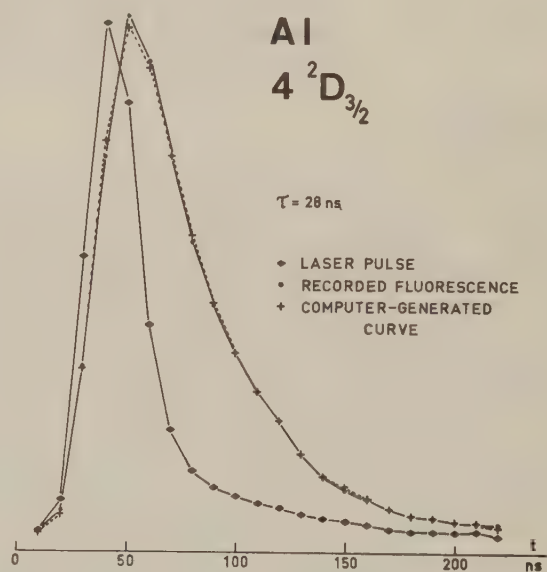


Fig. 2. Exciting laser pulse, recorded fluorescence and fitted computer generated curve corresponding to $\tau = 28\text{ ns}$ for the $4^2D_{3/2}$ -state

In the D -sequence we tried to distinguish between the $J=3/2$ and $J=5/2$ states. Unlike in indium [1] no difference in lifetime values was observed within the error bars. In addition to the discussed states the $6^2P_{3/2}$ level was measured using two-photon absorption. The laser light was then focused into the atomic beam. We also tried to measure in the $3s3p^2^4P$ term but due to good LS -coupling the signal was too weak to be recorded.

Table 1 gives our experimentally determined lifetime values. The table includes excitation wavelengths,

Table 1. Lifetime values for the $^2S_{1/2}$ and $^2D_{5/2,3/2}$ sequences of aluminum. Wavelengths for excitation from the $3p\ ^2P_{3/2}$ level are given (for the 2D states to the $J=5/2$ level). Methods of wavelength generation are also included. (KR=Kiton Red, R=Rhodamine, C=Coumarin, D=frequency doubling, 1 AS=1 anti-Stokes shift)

State	Lifetime [ns] This work	Lifetime [ns] Other works	Wavelength [Å]	Specifics of excitation	
				Nd: YAG system	Excimer system
$3s^24s^2S_{1/2}$		6.78 (6) ⁶ 6.0 ³	3,961.52		
5	24 (4)	16.8 ³	2,660.39	KR, D, 1AS	
6	52 (6)	37.0 ³	2,378.41	KR, D, 2AS	C470, D
7	99 (8)	69.6 ³	2,263.76	R 590, D, 2AS	C450, D
8	169 (17)	118 ³	2,204.64		C450, D
9	321 (40)		2,169.84	KR, D, 3AS	
10	462 (45)		2,147.41	KR, D, 3AS	
$3s^23d^2D_{5/2,3/2}$	16 (3)	13.7 (4) ⁷	3,092.72	R 640, D	
4	29 (4)		2,575.11	KR, D, 1AS	
5	15 (3)		2,373.13	KR, D, 2AS	C470, D
6	17 (3)		2,269.09	R 590, D, 2AS	C450, D
7	24 (4)		2,210.05		C450, D
8	30 (4)		2,174.03		C450, D
9	46 (5)		2,150.69	KR, D, 3AS	
10	59 (6)		2,134.74	KR, D, 3AS	
11	75 (6)		2,123.36	KR, D, 3AS	
$3s^26p^2P_{3/2}$	550 (50)		2·4,613.67		C470

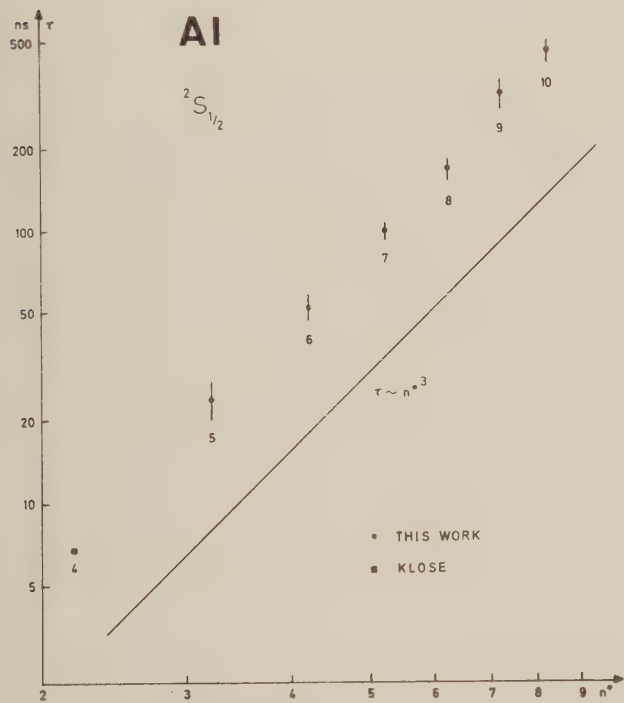


Fig. 3. The experimental lifetimes for the $3s^2ns^2S_{1/2}$ ($n=4-10$) states plotted versus the effective principal quantum number. Our error bars are indicated by the vertical lines

wavelength-generating system and values calculated or measured by other authors. The error bars in our measurements include both statistical scattering and estimated possible systematic errors in the used method.

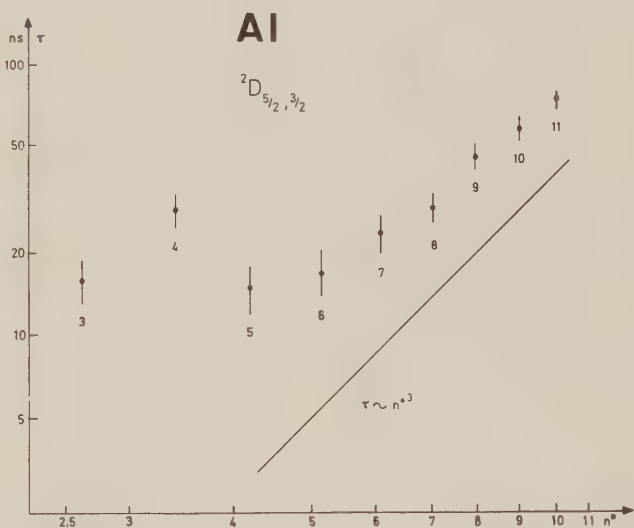


Fig. 4. The experimental lifetimes for the $3s^2nd^2D$ ($n=3-11$) states plotted versus the effective principal quantum number

4. Discussion

The experimental lifetimes from Table 1 are plotted in Figs.3 and 4. In an unperturbed Rydberg sequence the lifetimes are expected to scale with a power, close to three, of the effective principal quantum number. As can be seen in Fig. 3 this is the case for the $^2S_{1/2}$ sequence and from a fit $\tau=0.636 \cdot n_{\text{eff}}^{3.11}$ is obtained. Table 1 also includes lifetime values calculated with a semiempirical method [3]. The re-

sults from these calculations show good agreement with previously measured experimental oscillator strengths for lines combining with the ground levels. The calculated lifetime values are, however, about 30% lower than the experimental ones.

In contrast to the $^2S_{1/2}$ sequence the $^2D_{5/2, 3/2}$ sequence shows a strongly perturbed character. This was also the case in indium. Going from $n=4$ to 5 there is a decrease in lifetime but for higher states the values follow an n_{eff}^3 trend. This was supported by additional measurements on the $n=13$ state. The perturbation in the $^2D_{5/2, 3/2}$ sequence is caused by the $3s3p^2\ ^2D$ state. This state lies below the ionization limit and the perturbation is smeared out and it has been shown that the entire sequence is affected by configuration interaction [2].

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Hyperfine-Structure Study of the 7, 8 and 9²P_{3/2} States in ²³Na Using Quantum-Beat Spectroscopy

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Abstract

We have performed hyperfine-structure measurements on the 7, 8 and 9²P_{3/2} states in ²³Na using quantum-beat spectroscopy. The light from a pulsed dye laser, pumped by an excimer laser, was frequency-doubled and used to excite sodium atoms in an atomic beam. The fluorescence light was recorded by a transient digitizer and the transients were added in a digital memory and transferred to a mini-computer for calculations. For the magnetic-dipole and electric-quadrupole constants, *a* and *b*, we have obtained the following values:

$$\begin{aligned} 7^2P_{3/2}: & \quad a = 0.82(1) \text{ MHz}, \quad b = 0.13(3) \text{ MHz} \\ 8^2P_{3/2}: & \quad a = 0.535(15) \text{ MHz}, \quad b = 0.070(25) \text{ MHz} \\ 9^2P_{3/2}: & \quad a = 0.36(1) \text{ MHz}, \quad b = 0.045(15) \text{ MHz} \end{aligned}$$

1. Introduction

During the last few years pulsed tunable lasers with very high peak powers have been developed. This has made frequency mixing and doubling in non-linear crystals and frequency shifting in high-pressure gases quite efficient processes [1]. Tunable radiation at wavelengths down to 200 nm and below can be produced, which provides a very efficient way of selective population of highly excited atomic states using single-step excitation.

In this paper the hyperfine structure (hfs) of the 6–9²P_{3/2} states in ²³Na has been investigated using the quantum-beat method, employing a dye laser pumped by an excimer laser. From Fourier transforms of the time-resolved fluorescence decay curves, the magnetic-dipole and electric-quadrupole constants, *a* and *b*, were deduced. This experiment is an extension of an earlier quantum-beat investigation on sodium, where the 5 and 6²P_{3/2} states were studied using a pulsed nitrogen laser [2]. In the next section the experimental techniques are described in detail and in Section 3 the measurements and results are discussed. In the final section some conclusions are drawn.

2. Experimental technique

The experimental set-up is shown in Fig. 1. A pulsed dye-laser (Lambda Physik FL2002) with Coumarin 307 dye was pumped by an excimer laser (Lambda Physik EMG 102) with XeCl gas mixture. The dye-laser light was frequency doubled into the UV region using a KPB-crystal and a colour filter blocked the blue light. The energy of the 308 nm pump pulses was about 100 mJ using a 30 Hz repetition rate. The blue dye-

laser light had a pulsed energy of about 10 mJ. After frequency doubling UV pulses with an energy of approximately 0.1 mJ were obtained.

An atomic beam of ²³Na was created in a high-vacuum system. The sodium atoms were excited from the ²S_{1/2} ground state to the ²P_{3/2} states. The exciting pulses were 20 ns long and had a spectral width of about 0.1 Å. The laser light passed in and out of the vacuum system through long tubes in order to reduce stray light. The fluorescence on the exciting-light wavelength was analysed using a linear polarizer and an UV-interference filter and was detected with a photomultiplier tube (EMI 9558 BQ). Both excitation and detection were at right angles to the atomic beam.

After every laser pulse the fluorescence decay was captured by a transient digitizer (Biomation 8100). The transient digitizer was triggered by a photodiode detecting a small fraction of the dye-laser light. The time scale was calibrated by a radio frequency signal and each channel corresponded to 10 ns. The digitized data were transferred to a storage memory where a large number of transients could be added. For the lower states the fluorescence light was so strong that quantum-beats were obtained after a single laser pulse. However, to improve the signal to noise ratio, usually about one thousand transients were added before the signal was transferred to a mini-computer (PDP RT-11) and stored on a floppy disc.

3. Measurements and results

Our quantum-beat measurements were performed in the absence of external fields. The earth's magnetic field was compensated for by two Helmholtz-coil systems. ²³Na has a nuclear spin of 3/2 and a ²P_{3/2} state is split into four hfs levels with quantum number *F* = 3, 2, 1 and 0 (see Fig. 2). The interval between two hfs levels can be expressed in terms of magnetic-dipole and electric-quadrupole interaction constants *a* and *b* (see Fig. 2) [3].

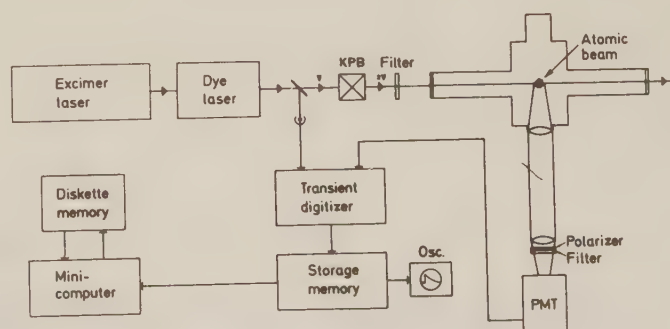


Fig. 1. Experimental set-up.

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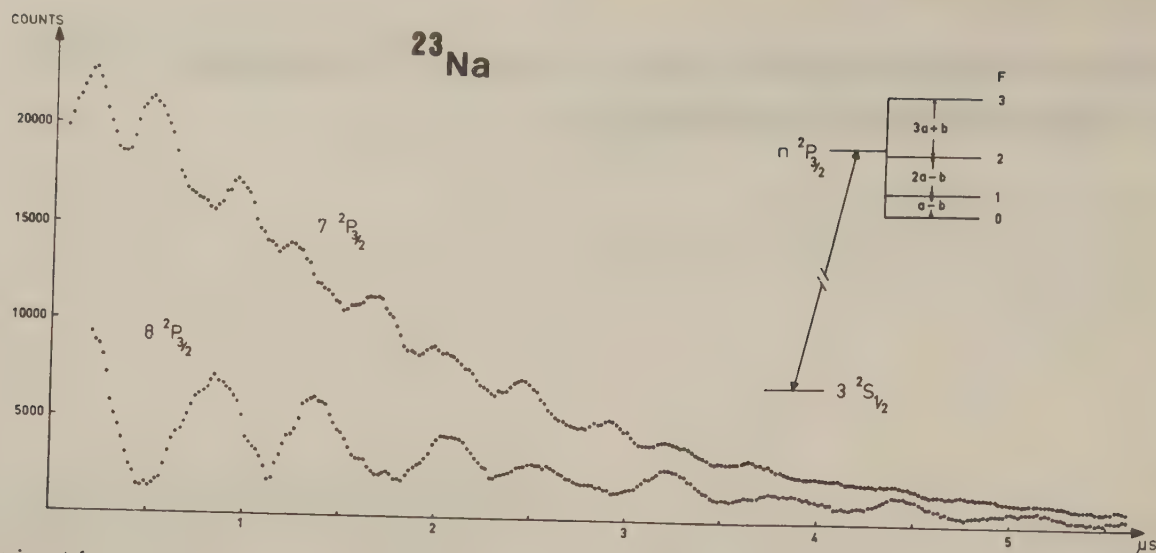


Fig. 2. Experimental quantum-beat curves for the $7\ 2P_{3/2}$ and the $8\ 2P_{3/2}$ states in ^{23}Na . The upper curve is recorded with the detection analyser parallel to the polarization of the laser light. The lower one is the differ-

ence between two curves recorded with orthogonal analyser settings. An excitation scheme showing the hfs levels is also included.

The selection rules for hfs quantum-beats $\Delta F = 2, 1$ yield five beat frequencies $5a$, $3a + b$, $3a - 2b$, $2a - b$ and $a - b$ [4]. The relative intensities of the beat components vary strongly and we could not detect them all.

The frequency-doubled light is linearly polarized. The polarizer on the detection side was set parallel or perpendicular to the incoming light. On turning the polarizer 90° the phase of the beats is shifted by 180° . For perpendicular polarizer setting the beat amplitudes are reduced by a factor of 2. On some occasions curves with different polarizer setting was subtracted, but normally only curves with parallel setting were used in the evaluations.

Two quantum-beat curves are shown in Fig. 2. The $7\ 2P_{3/2}$ curve is recorded with parallel polarizer setting while the $8\ 2P_{3/2}$ curve shows the result of subtraction with different polarizer settings. Subtracted curves have a signal with natural exponential damping, which can serve as an apodisation in the Fourier-

transform procedure. For curves with parallel polarizer setting we first calculated the exponential part of the decay. Each curve was then divided by its fitted exponential to get an undamped oscillatory signal. This gives a free choice of apodisation function in the Fourier transform procedure. A cosine-squared apodising function was used in a repeated Fourier-analysis scheme. By subtracting the most significant frequency in the Fourier spectrum and repeating the analysis we reduce frequency shifts in the weaker Fourier components [2]. In most evaluations the procedure was repeated three times. The first Fourier spectra from the two decay curves in Fig. 2 are shown in Fig. 3.

Using a large number of curves, the a and b factors for the $6-9\ 2P_{3/2}$ states were determined. The obtained values for the $6\ 2P_{3/2}$ state were entirely consistent with previous measurements [2]. The results for the other states are shown in Table 1 together with the excitation wavelengths. The experimental

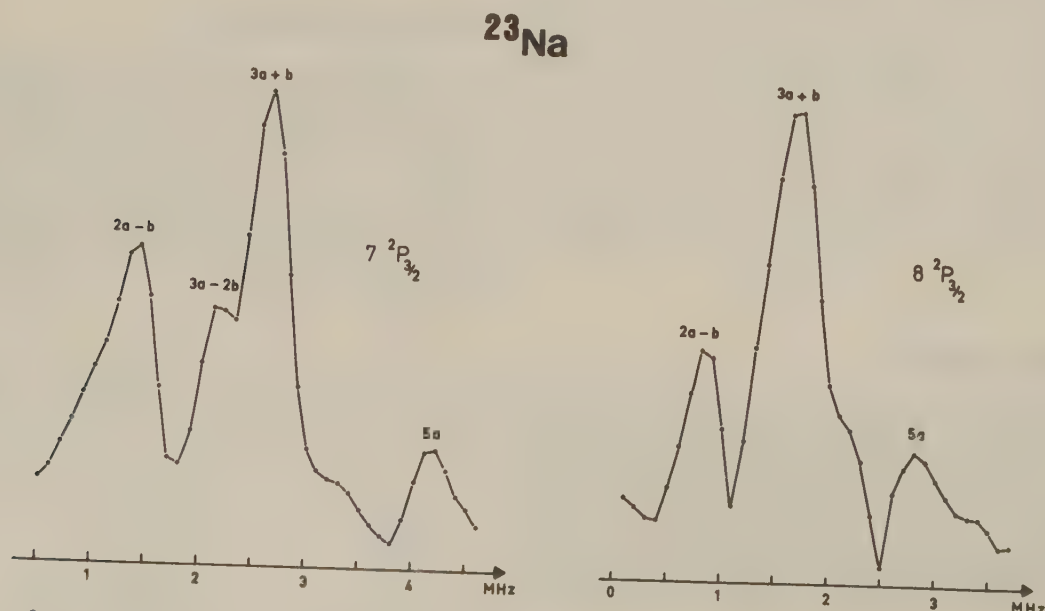


Fig. 3. Fourier transforms of the curves in Fig. 2. The left spectrum was obtained using $\cos^2$ -apodisation, while the right one has a natural exponential apodisation.

Table I. Experimental a and b factors, together with excitation wavelengths, for the studied ^{23}Na states

State	Excitation wavelength (Å)	a factor (MHz)	b factor (MHz)
$7^2P_{3/2}$	2594	0.82(1)	0.13(3)
$8^2P_{3/2}$	2544	0.535(15)	0.070(25)
$9^2P_{3/2}$	2512	0.36(1)	0.045(15)

factors were plotted vs. the effective principal quantum number in a log-log scale. As expected a straight line with a slope close to -3 was obtained.

4. Conclusions

The quantum-beat method employing high-energy pulsed lasers provides a very efficient way of high-resolution measurements in highly excited states. The laser tunability in the short wavelength region is especially interesting for spectroscopy on lighter

elements where the excitation energies usually are higher. Quantum-beat measurements on large energy separations require very fast detection systems. The demand for time-resolution is however much smaller if investigations are performed in magnetic fields close to a level-crossing as described in [5].

Acknowledgements

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Hyperfine Structure and Radiative Lifetimes in the $3s^2 np^2 P_{3/2}$ Sequence of ^{27}Al Using Time Resolved Laser Spectroscopy

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Using time-resolved laser spectroscopy we have determined the hyperfine structure in the $3s^2 np^2 P_{3/2}$ sequence of ^{27}Al ($I=5/2$). The magnetic-dipole interaction constants were found to scale with $n^{*-2.85}$ (n^* is the effective principal quantum number) in the investigated region $n=6-12$. Significant quadrupole interaction constants were obtained for $n=6-9$. The measured radiative lifetimes for $n=6-12$ scale with n^{*2} .

1. Introduction

The light group III element Al with ground configuration $3s^2 3p$ is well suited for testing theoretical calculations of quantities such as hyperfine interaction constants and radiative lifetimes. In a recent investigation [1] it was shown, that experimental lifetimes in the $3s^2 ns^2 S_{1/2}$ sequence scale very regularly as n^{*3} (n^* is the effective principal quantum number), whereas the $3s^2 nd^2 D_{5/2,3/2}$ sequence is strongly perturbed by admixture of the $3s3p^2^2D$ doubly excited state. This perturbation has been studied theoretically by Weiss [2] and by Lin [3]. In the present work, lifetime measurements have been extended to the $3s^2 np^2 P$ sequence, which for higher n -values may be influenced by black-body-radiation transitions [3,4].

With a single stable isotope ^{27}Al of nuclear spin $I=5/2$, aluminium is experimentally well suited for hyperfine structure (hfs) measurements. Still, information on hfs has been rather scarce. Apart from early atomic-beam magnetic resonance measurements on the ground configuration [6,7] and level-crossing measurements on the lowest $^2D_{5/2,3/2}$ states [8-11], investigations were only very recently performed for the $4s^2 S_{1/2}$ [12] and the $5p^2 P_{1/2}$ states [13]. With the quantum-beat spectroscopy (QBS) measurements reported in the present paper for the $6-12p^2 P_{3/2}$ states, hfs information has been substantially extended.

Beyond the Hartree-Fock model, polarization and correlation effects influence the hfs. The many-body

perturbation approach [14] is well suited for more accurate hfs calculations. In Ref. 13, calculations of the polarization effects were performed for low 2P levels of the group III elements Al, Ga and In. Here it was evident, that the non-included correlation effects are substantial, in particular for the ground configuration with three strongly overlapping outer electron orbitals (the outer s and p electrons have the same principal quantum number n). A full hfs calculation for the Al ground configuration is now being performed by Hörbäck and Lindgren [15]. The present measurements, which also include the quadrupole interaction, will allow interesting comparisons between the ground and the excited configurations what regards the influence of correlation.

2. Experimental Set-Up

In order to investigate high-lying $^2P_{3/2}$ states in aluminum, step-wise pulsed laser excitations via the shortlived $4s^2 S_{1/2}$ state were performed from the metastable $3p^2 P_{3/2}$ state of the ground configuration. The wavelength of the first excitation step is 396.2 nm, whereas the second step occurs in the range 555.7-455.0 nm for the $n=6-12$ states. Apart from the addition of a second dye laser, the experimental set-up is very similar to the one described in connection with QBS measurements on $^2P_{3/2}$ states in ^{23}Na [16]. A Lambda Physik EMG 102E ex-

cimer laser operating with XeCl at 308 nm was used at an energy level of about 100 mJ to pump two dye lasers. For the first step excitation, a homebuilt polyphenyl dye laser with a grating at grazing incidence was used [17]. The second step tunable radiation was obtained from a model FL 2002 dye laser. Coumarin 47, 153 and 307 dyes were employed. Pulse energies for the two lasers were 0.25 mJ and 1–10 mJ, respectively. The two beams were spatially and temporally overlapped and impinged on a beam of aluminium atoms formed in a vacuum system. The magnetic field in the scattering region could be controlled by Helmholtz coil systems. With a Karl Lambrecht double Fresnel rhomb polarization rotator the direction of polarization of the exciting beams could conveniently be rotated by 90° . The fluorescence light was detected by an EMI 9816 QB PMT through a linear polarizer. The optical transients were captured by a Biomation 8100 fast transient digitizer with a channel separation of 10 ns. Signal averaging was accomplished in a storage memory before data were transferred to a floppy disc and analysis on a PDP-11V03 minicomputer was performed.

3. Measurements and Results

In our lifetime measurements of the $6\text{--}12^2P$ states, separate determinations could be performed for the $^2P_{3/2}$ and $^2P_{1/2}$ states up to $n=11$. However, for $n=12$ the doublet does not resolve. In the spectro-

scopic literature only the 2P levels up to $7^2P_{3/2,1/2}$ have been specified [18]. The positions of the higher levels were calculated from the lower level quantum defects and no large deviations from expected positions were found. The lifetime experiments were performed in an external magnetic field of the order of 50 gauss and without employing a polarizer in the light detection. In this way any quantum beats are efficiently wiped out ensuring the detection of clean exponential decay curves. Normally, the time-resolved fluorescence was detected in the decay back to the $4s^2S_{1/2}$ state. However, sometimes the cascade decay via the shortlived $5s^2S_{1/2}$ state was observed yielding consistent results. In Fig. 1 three $^2P_{1/2}$ curves recorded in the direct decay are shown on a logarithmic scale. For each studied state typically 8 curves were recorded distributed over 3 different measurement occasions. The normal precautions to ensure unaffected lifetime results were taken [19]. Since we were not able to detect any significant differences in lifetimes for the two members of a doublet, only one value is given for each doublet. This is in marked contrast to the case of indium 2D states, where pronounced differences for the two doublet members were found [20]. The measured Al lifetime values are listed with estimated errors in Table 1 and the results are plotted on a log-log scale in Fig. 2. A straight line of a slope corresponding to the unperturbed n^3 dependence has been drawn in the figure. Clearly, the data do not follow this trend but rather the dependence $\tau = 27.4 \times n^{1.97}$ ns. It should be noted, that the 2P

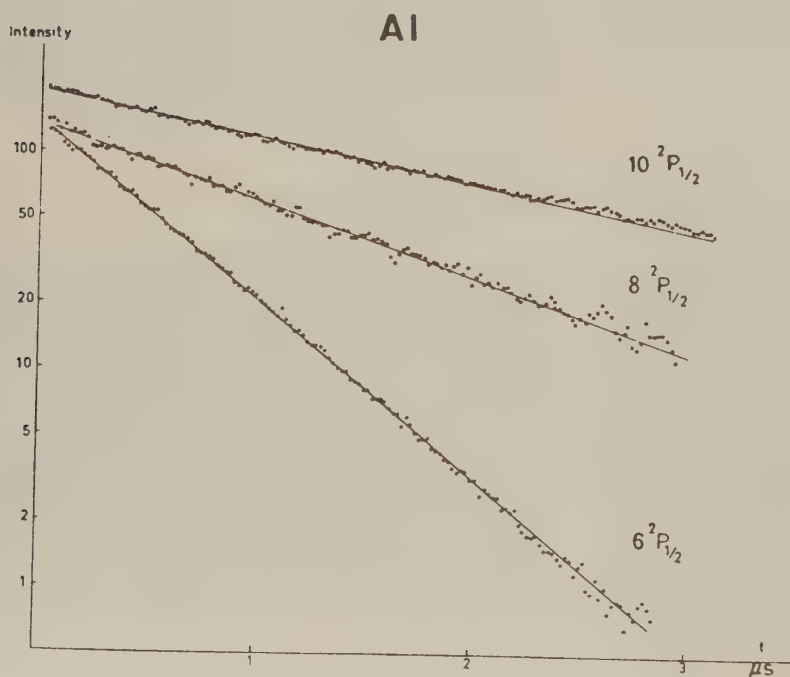


Fig. 1. Experimental decay curves for $^2P_{1/2}$ states of aluminum. For each curve about 1000 laser pulses have been used

Table 1. Measured radiative lifetimes for the $3s^2np^2P$ sequence of aluminium

State	Lifetime [ns]
$6^2P_{3/2,1/2}$	540(30)
7	920(70)
8	1,250(70)
9	1,520(90)
10	1,950(100)
11	2,410(170)
$12^2P_{3/2,1/2}$	2,880(260)

* This value is in good agreement with the value 550(50) ns obtained in two-photon absorption experiments [1]

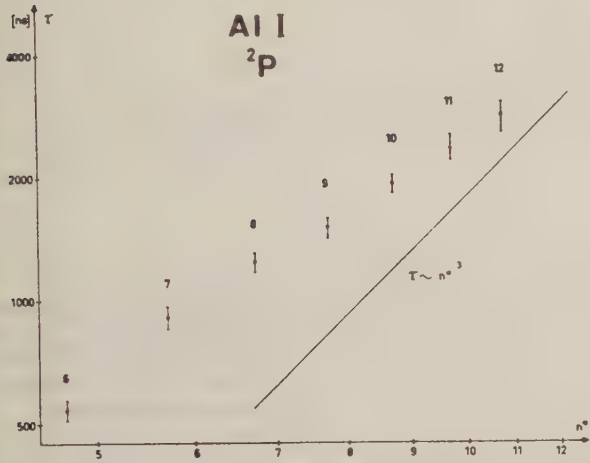


Fig. 2. Plot of radiative lifetimes of aluminum $^2P_{3/2,1/2}$ states versus n^* on a log-log scale. A line with a slope corresponding to an n^{*3} dependence is included

states are very long-lived compared to the 2S and 2D states [1]. Therefore they are very vulnerable to induced transitions due to the ambient black-body radiation [3,4]. The given lifetimes for higher n -values may be perturbed by the presence of the black-body radiation field. The values can be corrected to the case of zero kelvin using an approach corresponding to the one described in [5].

For the hyperfine quantum-beat measurements, external magnetic fields were eliminated by observing the disappearance of the otherwise very pronounced Zeeman quantum beats in the $6s6p^3P_1-6s^2^1S_0$ transition in ytterbium. The theoretical background for quantum-beat experiments has been presented by Haroche in [21]. Practical measurement and evaluation techniques are discussed in [22]. Selection rules for hyperfine structure quantum beats are $\Delta F=2,1$. With $J=3/2, I=5/2$ we then obtain the following possible beat frequencies [23]: $7a+0.35b$, $5a-1.25b$, $4a+0.8b$, $3a-0.45b$ and $2a-0.8b$. A maximum beat amplitude is obtained for parallel directions of the polarizers in the exciting and detected light. In Fig. 3 an example of a quantum beat recording for the $8p^2P_{3/2}$ state is shown. For perpendicular polarizer settings the phase of the beat signals is changed by 180° and the amplitude is reduced to its half value.

Thus, in order to isolate the beat structure, curves recorded with the different polarizer settings can be subtracted to eliminate the unmodulated signal while enhancing the beat structure. In Fig. 4 this type of curve is shown for the $10p^2P_{3/2}$ state. The

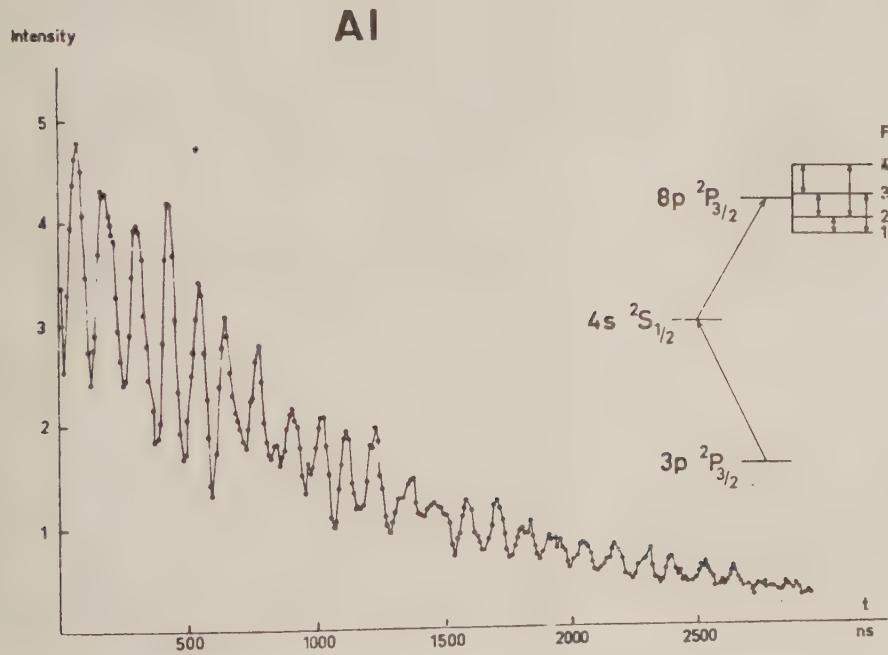


Fig. 3. Quantum-beat recording for the $8^2P_{3/2}$ state of ^{27}Al , obtained for parallel polarizer settings. About 2000 laser pulses were used for this curve

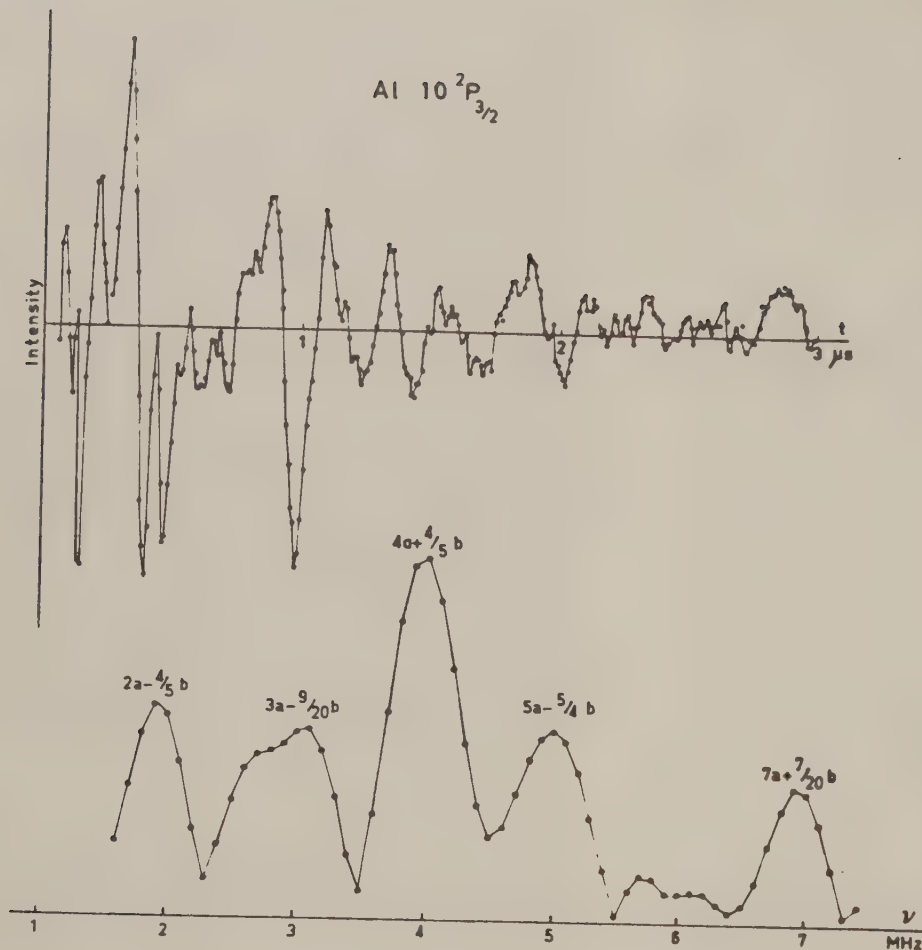


Fig. 4. Recording of quantum-beat for the $10\ ^2P_{3/2}$ state of ^{27}Al , where the exponential decay has been suppressed. A Fourier transform of the recording is also shown, featuring all five frequency components. A cosine-square apodization was used

Table 2. Experimental results for the magnetic-dipole interaction constant a and the electric-quadrupole interaction constant b for the $3s^2 np\ ^2P_{3/2}$ sequence in ^{27}Al

State	a [MHz]	b [MHz]
$3\ ^2P_{3/2}$	94.25(4) ^a	18.76(25) ^a
6	5.72(3)	0.47(5)
7	3.335(16)	0.31(3)
8	2.096(9)	0.180(15)
9	1.402(7)	0.150(30)
10	0.994(5)	
11	0.716(5)	
$12\ ^2P_{3/2}$	0.56(1)	

^a Ref. 6

corresponding Fourier transform obtained using a cosine-square apodisation [22] is included in the figure. For each of the studied $^2P_{3/2}$ states typically 6 curves were recorded, divided up on three different measurement occasions. In Table 2 results for the magnetic-dipole interaction constant a and the electric-quadrupole interaction constant b are given. The

measured a factors are plotted in Fig. 5 versus n^* on a log-log scale. As can be seen the points fall on a line, and in a fit we find $a = 477 \cdot n^{*-2.85}$. The spectroscopic resolution, which is proportional to $a/(1/2\pi\tau) \propto n^{*-0.88}$ decreases in the sequences at the same time as the signal to noise ratio decreases. This is reflected in the lower accuracy for the high n states. In particular, b factors can only be evaluated to a reasonable accuracy for the $n = 6-9$ states. In Ref. [13] we have performed some semiempirical calculations [24], illuminating the importance of the contact interaction in the magnetic hyperfine structure of 2P states of Al, Ga and In. Under simplifying assumptions it is possible to separate out the contact contribution to the measured a values for the $^2P_{3/2}$ and $^2P_{1/2}$ states of a doublet. The relevant radial parameter derived from the interaction constants, corrected for contact interaction, closely agrees with the parameter value derived from the experimental fine structure splitting [23]. Further, using that parameter essentially consistent values for the nuclear quadrupole moment Q are obtained using the mea-

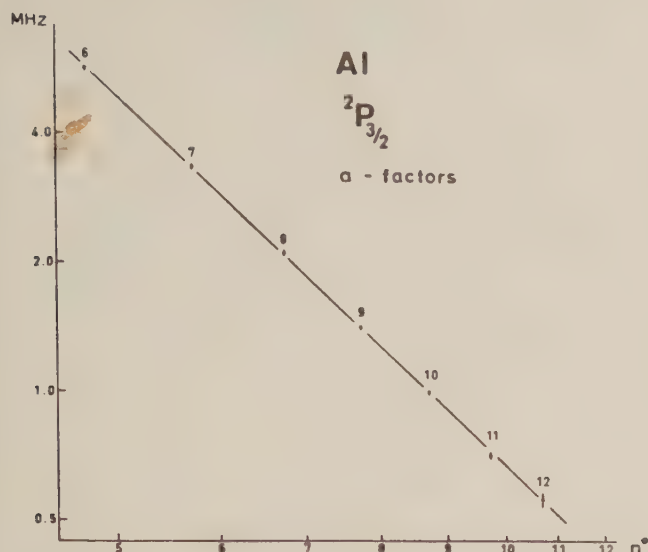


Fig. 5. Plot of magnetic-dipole interaction constants a for $^2P_{3/2}$ states of ^{27}Al versus n^* on a log-log scale. A fitted straight line of slope -2.85 is included

sured b factors for the investigated states. Since hfs information for the $6-9p^2P_{1/2}$ states is lacking, radial parameters can only be obtained from the fine structure splitting, only measured up to the 7^2P state [18]. However, such parameters were found to be reasonably reliable in Ref. [13]. Applying a Sternheimer correction factor of 0.94 [25] for the 3, 6 and $7^2P_{3/2}$ states we obtain the remarkably consistent results of 0.13, 0.14 and 0.15 barns, respectively, for the electric quadrupole moment of ^{27}Al . Thus, the semiempirical approach seems to work for Al as well as for Ga and In. Clearly, to gain a more detailed insight in the hfs interactions in group III elements, full many-body calculations must be performed. Such calculations are planned for the 2P sequence of Al [26], for which we also intend to perform further measurements, in particular to obtain the lacking data for the $^2P_{1/2}$ states.

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Hyperfine Structure and Isotope Shift of Highly Excited Barium-I States

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High-resolution laser-spectroscopy measurements of hyperfine structure and isotope shifts were performed for the $6snd\ ^1D_2$ sequence of Ba-I in the region $n=12-24$. Step-wise laser excitations of a collimated atomic beam were used. A strong influence on the hyperfine structure is observed at the perturbation at $n=14$, caused by interaction with the $5d7d$ configuration. Whereas the isotope shift for the even isotopes stays essentially constant with increasing n , the odd isotopes exhibit a strong change, indicating hyperfine-induced shifts.

1. Introduction

In alkaline-earth atoms with two electrons outside the closed shells, sequences of Rydberg states are frequently perturbed by doubly excited states of the same parity, converging towards a higher ionization limit. The knowledge of the energy-level structure for such systems has been considerably extended during the last few years through laser-spectroscopy measurements. E.g. for barium, studies have been performed by Bradley et al. [1], Rubbmark et al. [2] and Aymar et al. [3]. The observed energy-level structure has been interpreted by the last authors [3] and by Aymar and Robaux [4], using multi-channel quantum-defect theory. The configuration interaction is manifested through shifts in the level positions, but can also be studied in the strong perturbation of more subtle features of the states. For Ba the influence on Rydberg-state lifetimes due to interaction with $5d7d$ states have been investigated [5–8] as well as the corresponding perturbation in the Stark interaction [9, 6]. Also the Landé factors exhibit perturbations as shown by Grafström et al. [10]. In the present paper we show that hyperfine structure and isotope shifts sensitively probe the configuration interaction. High-resolution laser-spectroscopy experiments were performed for the $6snd\ ^1D_2$ sequence in the region $n=12-24$, covering the localized perturbation at $n=14$ caused by the $5d7d\ ^3F_2$ level. Measurements for the $n=7$ and 8

members of the sequence were previously reported by Jitschin and Meisel [11] using two-photon spectroscopy. Our experiments, which were performed using step-wise excitations of a collimated Ba atomic beam, show a strong resonant change in the hyperfine structure at the perturbed state. Whereas the isotope shift for the even isotopes stays essentially constant with increasing n , the odd isotopes exhibit a strong change indicating hyperfine-induced shifts. Preliminary results of the present investigation were presented in [12].

2. Experimental Arrangement

A scheme of the experimental arrangement used in the hyperfine-structure and isotope-shift measurements on Ba is given in Fig. 1. A multi-mode CW dye laser (Coherent Radiation CR 599), operating with the dye Rhodamine 110, was used to induce the $5,535\text{Å}$ resonance transition from the ground state $6s^2\ ^1S_0$ to the intermediate state $6s6p\ ^1P_1$. The lifetime of this state is about 8 ns [13] resulting in a natural radiation width of 20 MHz. In order to avoid problems due to the mode structure of the laser resulting in strong differences in population between the hyperfine levels of the 1P_1 state and to reduce population changes due to mode drifts and jitter, we vibrated the PZT-mounted folding mirror (tweeter) using a high-voltage oscillator. The fast

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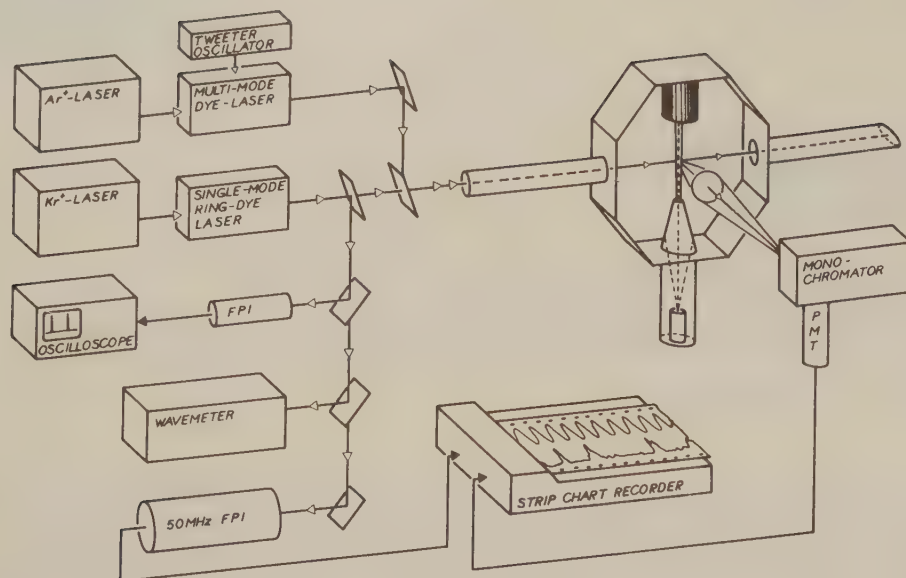


Fig. 1. Experimental set-up used in the hyperfine-structure and isotope-shift measurements

sweep of the modes back and forth resulted in an effectively "white" excitation over the $0.5 \text{ \AA}$ bandwidth of the laser. The second excitation step from the $6s6p^1P_1$ to the $6snd^1D_2$ states was induced with a Coherent Radiation model CR 699-21 single-mode ring-dye laser operating with Stilbene 3. The atoms to be studied were evaporated as an atomic beam in a vacuum system. Aperture stops along the beam were used for collimation and also helped suppress stray light from the oven. The two laser beams were superimposed using a dichroic mirror and were made to cross the atomic beam at right angles. Long entrance and exit tubes on the

vacuum system were used for the laser beams in order to avoid straylight. This was necessary because the detection followed at the same wavelength as the one used for the second-step excitation. The fluorescence scattering volume was imaged at the entrance slit of a 0.5 m Bausch and Lomb monochromator and was detected with an EMI 9558 BQ photomultiplier tube.

The wavelength of the single-mode laser was accurately set with a digital wavemeter [14] of a construction similar to that of Hall and Lee [15]. The frequency scan of the laser was monitored simultaneously with the recording of the fluorescence

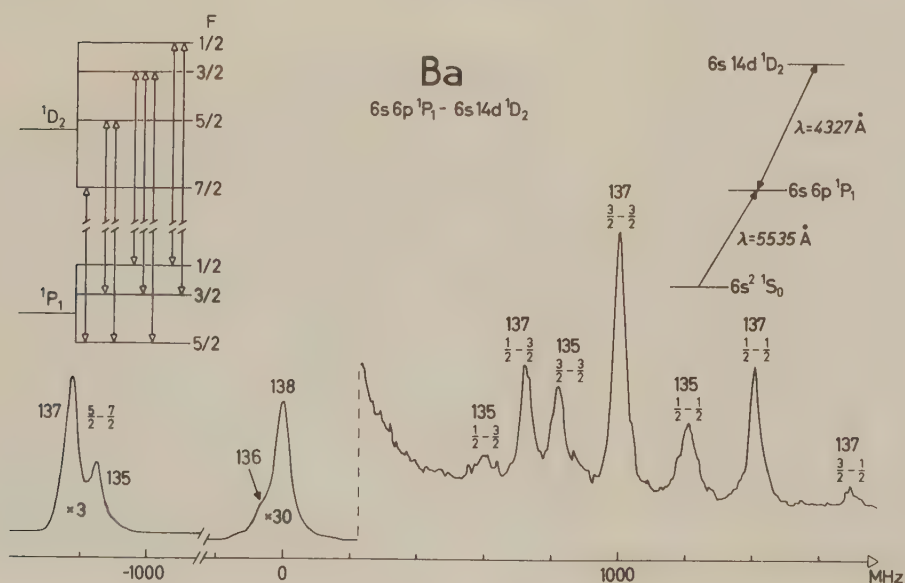


Fig. 2. Experimental curve for the $6s6p^1P_1 - 6s14d^1D_2$ transition at $4327 \text{ \AA}$. The excitation scheme and a schematic hyperfine transition diagram for ^{135}Ba and ^{137}Ba are included

spectrum using a fixed 50 MHz multipass Fabry-Pérot interferometer.

3. Measurements and Results

The diameter of the two laser beams, crossing the atomic beam was chosen to about 2 mm. It was observed that power broadening of the signal components easily occurs. We performed a careful investigation, attenuating the two laser beams independently by neutral-density filters, to ensure that light-shifts as well as power broadening were absent in the data recordings. We found that the width of our signals in excess of the natural linewidth due to the short-lived intermediate state was entirely due to residual Doppler broadening. The experimental linewidths were about 40 MHz.

A recording of the fluorescence light from the atomic beam during a single-mode laser scan of the $6s6p\ ^1P_1 - 6s14d\ ^1D_2$ transition at 4,327 Å is shown in Fig. 2. Components due to the isotopes with mass numbers 138, 137, 136 and 135 (natural abundances: 71.7, 11.3, 7.8 and 6.6 %) were observed. In Fig. 2 a transition diagram for the two odd isotopes is also included. Since both odd isotopes have nuclear spin $I=3/2$ and inverted hyperfine structures, the diagram schematically represents both of them. The evaluation of the magnetic-dipole constant a and the electric-quadrupole constant b was made from series of curves recorded on different occasions. The splittings of the $6s6p\ ^1P_1$ state as determined by Nowicki et al. [16] were used in the evaluation. For overlapping structures a computer program was employed to first determine the true positions of the peaks.

With the experimental linewidths and signal-to-noise ratio it was hard to evaluate accurate hyperfine constants for the less abundant odd isotope ^{135}Ba . As we are only interested in the electronic properties of the states reflected in the hyperfine structure and as the accuracy is too low to allow a determination of hyperfine anomalies we only give a and b factors for ^{137}Ba . However, isotope-shift values were calculated for all the isotopes 135, 136 and 137 with respect to the 138 isotope. For ^{135}Ba we then used the ratios $a_{137}/a_{135}=1.119$ and $b_{137}/b_{135}=1.537$ [17] to calculate the center of gravity from the strong ^{135}Ba peak positions. The experimental curves were compared with a computer-generated plot using the experimentally derived a , b and isotope-shift values. The very good agreement obtained warranted the used evaluation procedure. The experimental a and b factors for ^{137}Ba and the isotope shifts for the studied states are given in Table 1.

Table 1. Hyperfine-structure and isotope-shift results for Ba

$6snd\ ^1D_2$ n	a_{137} [MHz]	b_{137} [MHz]	IS in $6s6p\ ^1P_1 - 6snd\ ^1D_2$ line $\nu_{138} - \nu_A$ [MHz]		
			$A=135$	$A=136$	$A=137$
12	—	—	—	96(7)	—
13	-264(1)	10(7)	151(11)	98(4)	92(4)
14	-411(5)	83(16)	211(4)	69(9)	169(5)
15	-251(2)	12(9)	156(9)	107(6)	91(5)
16	-260(3)	4(8)	154(2)	101(4)	81(3)
17	-272(5)	-12(15)	121(11)	104(6)	40(6)
20	-346(2)	-29(18)	62(5)	102(6)	-22(2)
24	-530(2)	-128(9)	-193(16)	100(5)	-318(8)

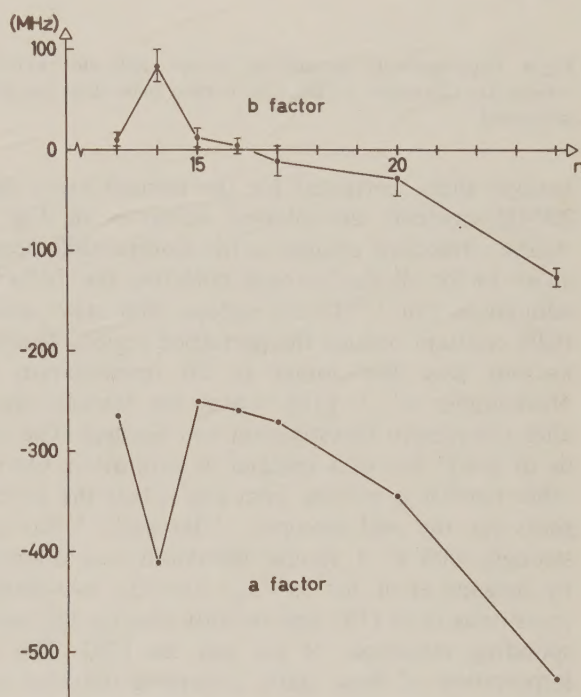


Fig. 3. Experimentally determined a and b factors for ^{137}Ba for $6snd\ ^1D_2$ states in the region $n=13-24$

4. Discussion

In Fig. 3 the hyperfine interaction constant data for ^{137}Ba are plotted as a function of n . The localized perturbation at $n=14$ due to the $5d7d\ ^3F_2$ state is manifested in a resonant change in both the a and b factors. Through the investigated region the interaction constants vary. Since the contribution to the hyperfine structure due to the outer electron is completely negligible the change reflects the gradual change in wavefunction composition.

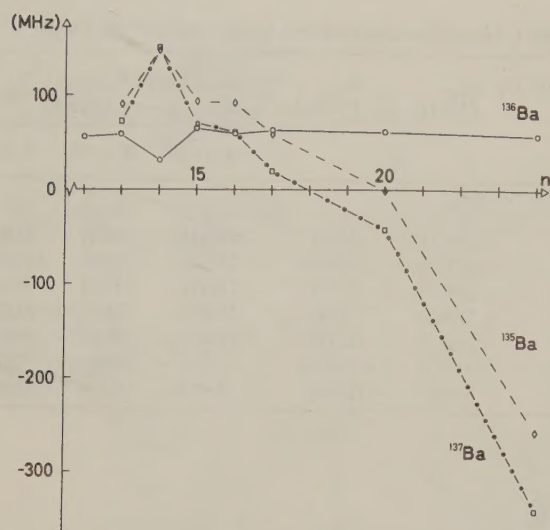


Fig. 4. Experimentally determined isotope shifts for $6s6p^1P_1 - 6snd^1D_2$ transitions of Ba. The normal mass shifts has been subtracted

Isotope shifts, corrected for the normal mass shift, 20 MHz/nucleon, are plotted versus n in Fig. 4. Again a resonant change in the isotope shift occurs at $n=14$ for all the isotopes reflecting the $5d7d^3F_2$ admixture. For ^{136}Ba the isotope shift stays essentially constant outside the perturbed region. This behaviour was also found in an investigation by Neukammer et al. [18], which we learned about after the present investigation was finished. The value of $|\psi(0)|^2$ has thus reached its saturation. On the other hand it is evident from Fig. 4 that the isotope shifts for the odd isotopes ^{135}Ba and ^{137}Ba vary strongly with n . A similar behaviour was observed by Beigang et al. for $5s^2^1S_0 - 5sns^1S_0$ two-photon transitions in Sr [19] and recently also for the corresponding sequences of Ca and Ba [20]. The interpretation of these shifts, occurring only for isotopes with non-zero nuclear spin, has been given in terms of hyperfine-induced singlet-triplet mixing [21, 20]. The situation is however more complicated for the 1D_2 states studied in the present paper because of the larger number of states and interactions that need to be considered and a detailed theoretical analysis is necessary.

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